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[Title of the Invention]  
Agricultural films

[Abstract]

[Solution]

Agricultural films of the present invention are of single layer structure or of multilayer structure consisting of two or more layers and are characterized in that they each have a layer of an ethylenic copolymer composition containing at a specific ratio an ethylene- $\alpha$ -olefin copolymer (A) composed of ethylene and an  $\alpha$ -olefin having 4 to 12 carbon atoms; a high-pressure-processed low-density polyethylene (B); an antifogging agent (C) which is a mixture of two or more kinds of nonionic surfactants, the mixture containing 15 to 50 % by weight of polyoxyethylene-

containing component; and a nonionic fluorine-containing surfactant (D) having a specific fluorine content. This copolymer (A) has a density, an MFR and an Mw/Mn in specific ranges respectively and also has a specific relationship between weight fraction of an n-decane-soluble component and a density at 23°C and also a relationship of  $B_1 \geq B_2$ , wherein  $B_1$  and  $B_2$  are an average branch number of the higher molecular weight part and that of the lower molecular weight part as determined by GPC-IR, respectively.

[Effect]

The above agricultural films have extremely excellent sustained antifogging properties and are also excellent in dust proofness, tenacity and extending workability.

[Scope of Claim for Patent]

[Claim 1]

An agricultural film having a layer of an ethylenic copolymer composition (1), the composition comprising:

[I] 100 parts by weight of ethylene- $\alpha$ -olefin copolymer (A) having a density of 0.905 to 0.935 g/cm<sup>3</sup> and a melt flow rate of 0.1 to 10 g/10 minutes to be obtained by copolymerization of ethylene with an  $\alpha$ -olefin having 4 to 12 carbon atoms, or 100 parts by weight of a mixture of the ethylene- $\alpha$ -olefin copolymer (A) and 40 % by weight or less of high-pressure-processed low-density polyethylene (B); and

[II] 0.05 to 5 parts by weight of antifogging agent (C) which is a mixture of two or more kinds of nonionic surfactants, the mixture containing 15 to 50 % by weight of polyoxyethylene-containing component; and

[III] 0.01 to 1 part by weight of nonionic fluorine-containing surfactant (D) having a fluorine content of 25 to 65 % by weight;

wherein the ethylene- $\alpha$ -olefin copolymer (A) or the mixture of the ethylene- $\alpha$ -olefin copolymer (A) and the high-pressure-processed low-density polyethylene (B):

(i) has a molecular weight distribution ( $M_w/M_n$  :  $M_w$  = weight average molecular weight,  $M_n$  = number average molecular weight) in the range of 1.5 to 5.0, as determined by GPC;

(ii) satisfies at 23°C the following relationship:

$$W < 80 \times \exp(-100(d-0.88)) + 0.1,$$

wherein  $W$  represents weight fraction (wt %) of an n-decane-soluble content; and  $d$  represents density; and

(iii) satisfies the relationship:  $B_1 \geq B_2$ ,

wherein  $B_1$  and  $B_2$  are an average branch number of the higher molecular weight part and that of the lower molecular weight part as determined by GPC-IR, respectively.

[Claim 2]

The agricultural film according to Claim 1, which is a single layer film of the ethylenic copolymer composition (1) containing the ethylene- $\alpha$ -olefin copolymer (A), the antifogging agent (C), the nonionic fluorine-containing surfactant (D) and, as necessary, the high-pressure-processed low-density polyethylene (B); wherein the film has:

(i) Elmendorf tear strength values of 90 kg/cm or more both in the MD (machine direction) and in the TD (transverse direction);

(ii) a dart impact strength of 450 g or more under a film thickness of 100  $\mu$ m; and

(iii) elongation at break values of 350 kg/cm<sup>2</sup> or more both in the MD and in the TD.

[Claim 3]

The agricultural film according to Claim 1 or 2, wherein the ethylene- $\alpha$ -olefin copolymer (A) is a copolymer of ethylene and an  $\alpha$ -olefin having 4 to 12 carbon atoms prepared using a metallocene catalyst.

[Claim 4]

The agricultural film according to any of Claims 1 to 3, wherein the polyoxyethylene-containing component constituting the antifogging agent (C) is represented by the formula  $(-\text{CH}_2\text{CH}_2\text{O}-)_n\text{H}$  (wherein  $n$  is 2 to 10) and has an HLB value of 5 to 12.

[Claim 5]

The agricultural film according to any of Claims 1 to 4, wherein the mixture of two or more kinds of nonionic surfactants constituting the antifogging agent (C) is a mixture of at least one kind of fatty acid ester selected from the group consisting of a glycerol fatty acid ester, a polyglycerol fatty acid ester and a sorbitan fatty acid ester; and at least one kind of polyoxyethylene-containing component selected from the group consisting of an adduct of a glycerol fatty acid ester with ethylene oxide, an adduct of a polyglycerol fatty acid ester with ethylene oxide and an adduct of sorbitan fatty acid ester with ethylene oxide.

[Claim 6]

The agricultural film according to Claim 5, wherein the glycerol fatty acid ester is glycerol monostearate.

[Claim 7]

The agricultural film according to Claim 5, wherein the polyglycerol fatty acid ester is diglycerol monostearate and/or diglycerol distearate.

[Claim 8]

The agricultural film according to Claim 5, wherein the sorbitan fatty acid ester is sorbitan monostearate and/or sorbitan distearate.

[Claim 9]

The agricultural film according to any of Claims 1 to 8, wherein the nonionic fluorine-containing surfactant (D) has a surface tension of 42 dyne/cm or less, as measured at 25°C in the form of 0.001 % aqueous solution.

[Claim 10]

The agricultural film according to any of Claims 1 to 9, wherein the ethylenic copolymer composition (1) contains an inorganic compound containing at least one atom selected from Mg, Ca, Al and Si in an amount of 1 to 20 parts by weight per 100 parts by weight of the total of the ethylene- $\alpha$ -olefin copolymer (A) and the high-pressure-processed low-density polyethylene (B).

[Claim 11]

The agricultural film according to any of Claims 1 to 10 having a thickness of 30 to 200  $\mu\text{m}$ .

[Claim 12]

The agricultural film according to Claim 1, which is a multilayer film formed by laminating:

[I] an outer layer of an ethylenic copolymer composition (2) containing an ethylene- $\alpha$ -olefin copolymer (E) having a density of 0.925 to 0.940  $\text{g/cm}^3$  and a melt flow rate of 0.1 to 10 g/10 minutes to be obtained by copolymerization of ethylene with an  $\alpha$ -olefin having 4 to 12

carbon atoms or containing a mixture of the ethylene- $\alpha$ -olefin copolymer (E) and 40 % by weight or less of high-pressure-processed low-density polyethylene (B);

[II] an intermediate layer of an ethylenic copolymer composition (3) containing 100 parts by weight of ethylene- $\alpha$ -olefin copolymer (F) having a density of 0.880 to 0.920 g/cm<sup>3</sup> and a melt flow rate of 0.1 to 10 g/10 minutes to be obtained by copolymerization of ethylene with an  $\alpha$ -olefin having 4 to 12 carbon atoms or containing 100 parts by weight of a mixture of the ethylene- $\alpha$ -olefin copolymer (F) and 40 % by weight or less of high-pressure-processed low-density polyethylene (B); and 0.05 to 5 parts by weight of antifogging agent (C) which is a mixture of two or more kinds of nonionic surfactants, the mixture containing 15 to 50 % by weight of polyoxyethylene-containing component; and

[III] an inner layer of the ethylenic copolymer composition (1) containing 100 parts by weight of the ethylene- $\alpha$ -olefin copolymer (A) having a density of 0.905 to 0.935 g/cm<sup>3</sup> and a melt flow rate of 0.1 to 10 g/10 minutes to be obtained by copolymerization of ethylene with  $\alpha$ -olefin having 4 to 12 carbon atoms or containing 100 parts by weight of a mixture of the ethylene- $\alpha$ -olefin copolymer (A) and 40 % by weight or less of pressure-processed low-density polyethylene (B), and 0.05 to 5 parts by weight of antifogging agent (C) which is a mixture of two or more kinds of nonionic surfactants, the mixture containing 15 to 50 % by weight of polyoxyethylene-containing component;

wherein among the ethylenic copolymer compositions (1), (2) and (3), at least the ethylenic copolymer composition (1) contains 0.01 to 1 part by weight of nonionic fluorine-containing surfactant (D) having a fluorine content of 25 to 65 % by weight; the density ( $d_E$  or  $d_{EB}$ ) of the ethylene- $\alpha$ -

olefin copolymer (E) or of the mixture of the ethylene- $\alpha$ -olefin copolymer (E) and the pressure-processed low-density polyethylene (B) forming the outer layer and the density ( $d_A$  or  $d_{AB}$ ) of the ethylene- $\alpha$ -olefin copolymer (A) or of the mixture of the ethylene- $\alpha$ -olefin copolymer (A) and the pressure-processed low-density polyethylene (B) satisfy the following relationship:  $d_E$  or  $d_{EB} \geq d_A$  or  $d_{AB}$ .

[Claim 13]

The agricultural film according to Claim 12, wherein the ethylene- $\alpha$ -olefin copolymer (E) or the mixture of the ethylene- $\alpha$ -olefin copolymer (E) and the high-pressure-processed low-density polyethylene (B) forming the outer layer; and the ethylene- $\alpha$ -olefin copolymer (F) or the mixture of the ethylene- $\alpha$ -olefin copolymer (F) and the high-pressure-processed low-density polyethylene (B) forming the intermediate layer:

(i) each have a molecular weight distribution ( $M_w/M_n$  :  $M_w$  = weight average molecular weight,  $M_n$  = number average molecular weight) in the range of 1.5 to 5.0, as determined by GPC;

(ii) satisfy at 23°C the following relationship:

$$W < 80 \times \exp(-100(d-0.88)) + 0.1,$$

wherein W represents weight fraction (wt %) of an n-decane-soluble content; and d represents density; and

(iii) satisfy the relationship:  $B_1 \geq B_2$ ,

wherein  $B_1$  and  $B_2$  are an average branch number of the higher molecular weight part and that of the lower molecular weight part as determined by GPC-IR, respectively.

[Claim 14]

The agricultural film according to Claim 12 or 13, wherein the ethylene- $\alpha$ -olefin copolymers (E) and (F) each are a copolymer of ethylene and an  $\alpha$ -olefin having 4 to 12 carbon atoms prepared using a metallocene catalyst.

[Claim 15]

The agricultural film according to any of Claims 12 to 14, wherein the antifogging agent (C) is contained in the ethylenic copolymer composition (2) in an amount of 0.05 to 5 parts by weight per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (E) and the high-pressure-processed low-density polyethylene (B).

[Claim 16]

The agricultural film according to any of Claims 12 to 15, wherein the polyoxyethylene-containing component constituting the antifogging agent (C) is represented by the formula  $(-\text{CH}_2\text{CH}_2\text{O}-)_n\text{H}$  (wherein  $n$  is 2 to 10) and has an HLB value of 5 to 12.

[Claim 17]

The agricultural film according to any of Claims 12 to 16, wherein the mixture of two or more kinds of nonionic surfactants constituting the antifogging agent (C) is a mixture of at least one kind of fatty acid ester selected from the group consisting of a glycerol fatty acid ester, a polyglycerol fatty acid ester and a sorbitan fatty acid ester; and at least one kind of polyoxyethylene-containing component selected from the group consisting of an adduct of a glycerol fatty acid ester with ethylene oxide, an adduct of a polyglycerol fatty acid ester with ethylene oxide and an adduct of sorbitan fatty acid ester with ethylene oxide.

[Claim 18]

The agricultural film according to Claim 17, wherein the glycerol fatty acid ester is glycerol monostearate.

[Claim 19]

The agricultural film according to Claim 17, wherein the polyglycerol fatty acid ester is diglycerol monostearate and/or diglycerol distearate.

[Claim 20]

The agricultural film according to Claim 17, wherein the sorbitan fatty acid ester is sorbitan monostearate and/or sorbitan distearate.

[Claim 21]

The agricultural film according to any of Claims 12 to 20, wherein the nonionic fluorine-containing surfactant (D) has a surface tension of 42 dyne/cm or less, as measured at 25°C in the form of 0.001 % aqueous solution.

[Claim 22]

The agricultural film according to any of Claims 12 to 21, wherein the ethylenic copolymer composition (2) contains an inorganic compound containing at least one atom selected from Mg, Ca, Al and Si in an amount of 1 to 20 parts by weight per 100 parts by weight of the total of the ethylene- $\alpha$ -olefin copolymer (E) and the high-pressure-processed low-density polyethylene (B).

[Claim 23]

The agricultural film according to any of Claims 12 to 22, wherein the ethylenic copolymer composition (3) contains an inorganic compound containing at least one atom selected

from Mg, Ca, Al and Si in an amount of 1 to 20 parts by weight per 100 parts by weight of the total of the ethylene- $\alpha$ -olefin copolymer (E) and the high-pressure-processed low-density polyethylene (B).

[Claim 24]

The agricultural film according to any of Claims 12 to 23, wherein the outer layer [I], the intermediate layer [II] and the inner layer [III] have a thickness ratio (outer layer [I]:intermediate layer [II]:inner layer [III]) of 0.2-4/1-10/1, and the total thickness of these layers is 30 to 200  $\mu\text{m}$ .

[Claim 25]

The agricultural film according to any of Claims 12 to 24, wherein the film has:

(i) Elmendorf tear strength values of 90 kg/cm or more both in the MD (machine direction) and in the TD (transverse direction); and

(ii) a dart impact strength of 900 g or more under a film thickness of 100  $\mu\text{m}$ ; and

(iii) elongation at break values of 350 kg/cm<sup>2</sup> or more both in the MD and in the TD.

[Detailed Description of the Invention]

[0001]

[Technical Field of the Invention]

The present invention relates to an agricultural film.

[0002]

[Technical Background of the Invention]

Growing plants in plastic greenhouses and plastic

tunnels or with mulching for the purpose of agricultural forcing culture generally uses large amounts of agricultural films made of various types of thermoplastic resins as covering materials. Such agricultural films include, for example, polyvinyl chloride films, ethylene-vinyl acetate copolymer films and polyethylene films. Among others, polyvinyl chloride films are most frequently used because of their excellent thermo-keeping properties, transparency, tenacity (mechanical strength), etc.

[0003]

However, the polyvinyl chloride film involves problems, for example, that it has rather poor dust proofness and poor extending workability and develops a toxic gas when it is incinerated. Here, the expression "thermo-keeping properties" means the performance of absorbing and reflecting radiation emitted during the night from the earth heated during the daytime by absorbing solar energy to keep the temperature (air temperature and earth temperature) in the plastic greenhouses or tunnels.

[0004]

Meanwhile, the expression "dust proofness" means the performance of inhibiting decline in the transparency of the film to be caused by deposition of dust and the like after use for a certain period. Further, the expression "extending workability" means handleability of the film evaluated depending on the tackiness.

[0005]

Meanwhile, the film based on an ethylene-vinyl acetate copolymer is superior to the polyvinyl chloride film in

terms of waste treatment but has poor tenacity and are insufficient in extending workability and dust proofness.

[0006]

Meanwhile, if a polyethylene film is used for agricultural uses, this film involves problems that, although it may have improved extending workability and dustproofness compared with the film based on ethylene-vinyl acetate copolymer, the degree of improvement is still insufficient and that it has poor tenacity compared with linear ethylene- $\alpha$ -olefin copolymer films.

[0007]

Further, there are polyolefin resin films for agricultural uses whose thermo-keeping properties are improved by additives, for example, a covering film prepared by adding silicon oxide to a low-density polyethylene or an ethylene-vinyl acetate copolymer.

[0008]

However, such agricultural films have insufficient tenacity and transparency compared with the polyvinyl chloride film, although they have improved thermo-keeping properties compared with the polyethylene resin film.

[0009]

With a view to improving tenacity of such polyethylene resin films, agricultural films utilizing tenacity of linear low-density polyethylenes have recently been devised. For example, Japanese Unexamined Patent Publication (KOKAI) No. Sho 58-160146 proposes an agricultural multilayer film having excellent antifogging properties formed by laminating a substrate layer based on a linear low-density polyethylene and a layer of a polyethylene resin containing a surfactant,

prepared by the conventional procedure. This agricultural multilayer film has improved tenacity but has extremely poor thermo-keeping properties compared with the polyvinyl chloride film since the former does not discuss utilization of a resin having a higher thermo-keeping properties or additives.

[0010]

Meanwhile, Japanese Unexamined Patent Publication (KOKAI) No. Hei 1-182037 proposes an agricultural multilayer (laminated) film formed by an outer layer containing a polyethylene resin based on a linear ethylene- $\alpha$ -olefin copolymer and an inorganic compound, and an inner layer containing a polyethylene resin based on an ethylene-vinyl acetate copolymer and an inorganic compound and an antifogging agent. Japanese Unexamined Patent Publication (KOKAI) No. Hei 1-182037 describes an agricultural multilayer film that has extremely excellent tenacity in addition to excellent thermo-keeping properties, dust proofness, extending workability, transparency and weathering resistance.

[0011]

However, in the agricultural multilayer film as described above, there is still room for improvement in terms of sustained antifogging properties, dust proofness, tenacity, etc. Therefore, there is desired an advent of agricultural film, which undergoes minor drop in sustained antifogging properties with time and which has excellent dust proofness and tenacity, compared with the conventional agricultural films as described above (including both the single layer films and the multilayer films).

[0012]

[Object of the Invention]

It is an objective of the present invention to provide an agricultural film, which undergoes minor drop in sustained antifogging properties with time and which has excellent dust proofness and tenacity, compared with the conventional agricultural films.

[0013]

[Summary of the Invention]

The agricultural film according to the present invention has a layer of an ethylenic copolymer composition (1), the composition containing:

[I] 100 parts by weight of ethylene- $\alpha$ -olefin copolymer (A) having a density of 0.905 to 0.935 g/cm<sup>3</sup> and a melt flow rate of 0.1 to 10 g/10 minutes to be obtained by copolymerization of ethylene with an  $\alpha$ -olefin having 4 to 12 carbon atoms, or 100 parts by weight of a mixture of the ethylene- $\alpha$ -olefin copolymer (A) and 40 % by weight or less of high-pressure-processed low-density polyethylene (B); and

[II] 0.05 to 5 parts by weight of antifogging agent (C) which is a mixture of two or more kinds of nonionic surfactants, the mixture containing 15 to 50 % by weight of polyoxyethylene-containing component; and

[III] 0.01 to 1 part by weight of nonionic fluorine-containing surfactant (D) having a fluorine content of 25 to 65 % by weight;

wherein the ethylene- $\alpha$ -olefin copolymer (A) or the mixture of the ethylene- $\alpha$ -olefin copolymer (A) and the high-pressure-processed low-density polyethylene (B):

(i) has a molecular weight distribution ( $M_w/M_n$  :  $M_w$  = weight average molecular weight,  $M_n$  = number average molecular weight) in the range of 1.5 to 5.0, as determined by GPC;

(ii) satisfies at 23°C the following relationship:

$$W < 80 \times \exp(-100(d-0.88)) + 0.1,$$

wherein W represents weight fraction (wt %) of an n-decane-soluble content; and d represents density; and

(iii) satisfies the relationship:  $B_1 \geq B_2$ ,

wherein  $B_1$  and  $B_2$  are an average branch number of the higher molecular weight part and that of the lower molecular weight part as determined by GPC-IR, respectively.

[0014]

The agricultural film according to the present invention may be of a single layer structure or a multilayer structure having two or more layers. A preferred single-layer structure agricultural film according to the present invention is a single layer film of a layer of the ethylenic copolymer composition (1) containing the ethylene- $\alpha$ -olefin copolymer (A), the antifogging agent (C), the nonionic fluorine-containing surfactant (D) and, as necessary, the high-pressure-processed low-density polyethylene (B); wherein the film has

(i) Elmendorf tear strength values of 90 kg/cm or more both in the MD (machine direction) and in the TD (transverse direction);

(ii) a dart impact strength of 450 g or more under a film thickness of 100  $\mu\text{m}$ ; and

(iii) elongation at break values of 350 kg/cm<sup>2</sup> or more both in the MD and in the TD.

[0015]

Further, a preferred multilayer agricultural film according to the present invention is formed by laminating:

[I] an outer layer of an ethylenic copolymer composition (2) containing an ethylene- $\alpha$ -olefin copolymer

(E) having a density of 0.925 to 0.940 g/cm<sup>3</sup> and a melt flow rate of 0.1 to 10 g/10 minutes to be obtained by copolymerization of ethylene with an  $\alpha$ -olefin having 4 to 12 carbon atoms or containing a mixture of the ethylene- $\alpha$ -olefin copolymer (E) and 40 % by weight or less of high-pressure-processed low-density polyethylene (B);

[II] an intermediate layer of an ethylenic copolymer composition (3) containing 100 parts by weight of ethylene- $\alpha$ -olefin copolymer (F) having a density of 0.880 to 0.920 g/cm<sup>3</sup> and a melt flow rate of 0.1 to 10 g/10 minutes to be obtained by copolymerization of ethylene with an  $\alpha$ -olefin having 4 to 12 carbon atoms or containing 100 parts by weight of a mixture of the ethylene- $\alpha$ -olefin copolymer (F) and 40 % by weight or less of high-pressure-processed low-density polyethylene (B); and 0.05 to 5 parts by weight of antifogging agent (C) which is a mixture of two or more kinds of nonionic surfactants, the mixture containing 15 to 50 % by weight of polyoxyethylene-containing component; and

[III] an inner layer of the ethylenic copolymer composition (1) containing 100 parts by weight of the ethylene- $\alpha$ -olefin copolymer (A) having a density of 0.905 to 0.935 g/cm<sup>3</sup> and a melt flow rate of 0.1 to 10 g/10 minutes to be obtained by copolymerization of ethylene with  $\alpha$ -olefin having 4 to 12 carbon atoms or containing 100 parts by weight of a mixture of the ethylene- $\alpha$ -olefin copolymer (A) and 40 % by weight or less of pressure-processed low-density polyethylene (B), and 0.05 to 5 parts by weight of antifogging agent (C) which is a mixture of two or more kinds of nonionic surfactants, the mixture containing 15 to 50 % by weight of polyoxyethylene-containing component;

wherein among the ethylenic copolymer compositions (1), (2) and (3), at least the ethylenic copolymer composition

(1) contains 0.01 to 1 part by weight of nonionic fluorine-containing surfactant (D) having a fluorine content of 25 to 65 % by weight; the density ( $d_E$  or  $d_{EB}$ ) of the ethylene- $\alpha$ -olefin copolymer (E) or of the mixture of the ethylene- $\alpha$ -olefin copolymer (E) and the pressure-processed low-density polyethylene (B) forming the outer layer and the density ( $d_A$  or  $d_{AB}$ ) of the ethylene- $\alpha$ -olefin copolymer (A) or of the mixture of the ethylene- $\alpha$ -olefin copolymer (A) and the pressure-processed low-density polyethylene (B) satisfy the following relationship:  $d_E$  or  $d_{EB} \geq d_A$  or  $d_{AB}$ .

[0016]

Meanwhile, the resins constituting the outer layer, the intermediate layer and the inner layer respectively:

(i) each have a molecular weight distribution ( $M_w/M_n$  :  $M_w$  = weight average molecular weight,  $M_n$  = number average molecular weight) in the range of 1.5 to 5.0 determined by GPC;

(ii) satisfy at 23°C the following relationship:

$$W < 80 \times \exp(-100(d-0.88)) + 0.1,$$

wherein  $W$  represents weight fraction (wt %) of an n-decane-soluble content; and  $d$  represents density; and

(iii) satisfy the relationship:  $B_1 \geq B_2$ ,

wherein  $B_1$  and  $B_2$  are an average branch number of the higher molecular weight part and that of the lower molecular weight part as determined by GPC-IR, respectively. The expression "resins" referred to here means an ethylene- $\alpha$ -olefin copolymer or a copolymer of an ethylene- $\alpha$ -olefin copolymer with a high-pressure-processed low-density polyethylene.

[0017]

Further, the agricultural film having the multilayer structure as described above preferably has:

(i) Elmendorf tear strength values of 90 kg/cm or more both in the MD (machine direction) and in the TD (transverse direction);

(ii) a dart impact strength of 900 g or more under a film thickness of 100  $\mu\text{m}$ ; and

(iii) elongation at break values of 350 kg/cm<sup>2</sup> or more both in the MD and in the TD.

[0018]

[Detailed Description of the Invention]

The agricultural film according to the present invention will be described below specifically. The agricultural film according to the present invention has a layer of an ethylenic copolymer composition (1) containing an ethylene- $\alpha$ -olefin copolymer (A), an antifogging agent (C), a nonionic fluorine-containing surfactant (D) and, as necessary, a high-pressure-processed low-density polyethylene (B). The agricultural film according to the present invention may be a single layer structure film (single-layer film) made of such layer or a film having a multilayer structure (multilayer film) containing two or more such layers.

[0019]

[Ethylene- $\alpha$ -olefin copolymer (A)]

The ethylene- $\alpha$ -olefin copolymer (A) constituting the ethylenic copolymer composition (1) employed in the present invention is one containing ethylene and an  $\alpha$ -olefin having 4 to 12 carbon atoms.

[0020]

$\alpha$ -Olefins having 4 to 12 carbon atoms employable for copolymerization with ethylene include typically 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene, 1-octene, 1-decene and 1-dodecene, preferably  $\alpha$ -olefins having 4 to 12 carbon atoms, and particularly preferably  $\alpha$ -olefins having 4 to 6 carbon atoms.

[0021]

In the present invention, such ethylene- $\alpha$ -olefin copolymers (A) can be used singly or as a combination of two or more kinds of them. Ethylene- $\alpha$ -olefin copolymers (A) to be employed according to the present invention each desirably have a structural unit derived from ethylene in an amount of 50 % by weight or more and less than 100 % by weight, preferably 55 to 99 % by weight, more preferably 65 to 98 % by weight, and particularly preferably 70 to 96 % by weight, and a structural unit derived from  $\alpha$ -olefin having 4 to 12 carbon atoms in an amount of 50 % by weight or less, preferably 1 to 45 % by weight, more preferably 2 to 35 % by weight, particularly preferably 4 to 30 % by weight.

[0022]

The composition of the ethylene- $\alpha$ -olefin copolymer is usually decided under measurement of  $^{13}\text{C}$ -NMR spectrum of a sample prepared by dissolving about 200 mg of the copolymer in 1 ml of hexachlorobutadiene homogeneously in a 10 mm-diameter test tube under the following conditions:

measurement temperature:	120°C
measurement frequency:	25.05 MHz
spectrum width:	1500 OHZ
pulse repetition period:	4.2 sec.
pulse width:	6 $\mu$ sec.

[0023]

Meanwhile, the ethylene- $\alpha$ -olefin copolymer (A) has a density in the range of 0.905 to 0.935 g/cm<sup>3</sup>, preferably 0.908 to 0.935 g/cm<sup>3</sup>, more preferably 0.910 to 0.930 g/cm<sup>3</sup>, and particularly preferably 0.925 to 0.930 g/cm<sup>3</sup>.

[0024]

Here, the density is determined as follows: A strand of the ethylene- $\alpha$ -olefin copolymer (A) obtained in the measurement of melt flow rate (MFR) under a load of 2.16 kg is subjected to heat treatment at 120°C for one hour and then cooled gradually to room temperature over one hour, followed by measurement of density using a density-gradient tube.

[0025]

The ethylene- $\alpha$ -olefin copolymer (A) has a melt flow rate (MFR: ASTM D 1238-65T, 190°C, load 2.16 kg) in the range of 0.1 to 10 g/10 minutes, preferably 0.1 to 5 g/10 minutes, and more preferably 0.5 to 2 g/10 minutes.

[0026]

This ethylene- $\alpha$ -olefin copolymer (A) has a molecular weight distribution (Mw/Mn : Mw = weight average molecular weight; Mn = number average molecular weight) of 1.5 to 5.0, preferably 1.8 to 3.5, more preferably 2.0 to 3.0, as determined by GPC.

[0027]

Here, the molecular weight distribution (Mw/Mn) was determined using a GPC-150C (Millipore) as follows:

A separation column (GNH HT, TSK) having a diameter of 72 mm and a length of 600 mm was used. The column temperature was 140°C. o-Dichlorobenzene (Wako Pure Chemical Ind., Ltd.) was used as a mobile phase, and 0.025 % by weight of BHT (Takeda Chemical Industries, Ltd.) was used as an antioxidant. The mobile phase was moved at a rate of 1.0 ml/min. The concentration of the sample was 0.1 wt %. The sample was applied in an amount of 500 µl. A differential refractometer was used as a detector. As reference polystyrenes were used those of Toso Co. Ltd. having molecular weights of  $M_w < 1000$  and  $M_w > 4 \times 10^5$  respectively and those of Pressure Chemical having molecular weights of  $1000 < M_w < 4 \times 10^5$ .

[0028]

The ethylene- $\alpha$ -olefin copolymer (A) satisfies the following relationship:

[0029]

$$W < 80 \times \exp(-100(d-0.88)) + 0.1,$$

wherein W represents weight fraction (wt %) of an n-decane-soluble content; and d represents density (d). Here, the n-decane-soluble content of the ethylene- $\alpha$ -olefin copolymer (the smaller the soluble content, the narrower the compositional distribution) is determined by adding about 3 g of ethylene- $\alpha$ -olefin copolymer to 450 ml of n-decane to be dissolved therein at 145°C, and then cooling the solution to 23°C, followed by filtration to remove n-decane-insoluble content and recovering of the n-decane-soluble content from the filtrate.

[0030]

Further, the ethylene- $\alpha$ -olefin copolymer desirably satisfies the following relationship:

$$T_m < 400 \times d - 250,$$

$$\text{preferably } T_m < 450 \times d - 297,$$

$$\text{more preferably } T_m < 500 \times d - 344,$$

$$\text{particularly preferably } T_m < 550 \times d - 391,$$

wherein  $T_m$  represents the maximum peak temperature ( $^{\circ}\text{C}$ ) of the endothermic curve determined using a differential scanning calorimeter; and  $d$  represents density ( $\text{g}/\text{cm}^3$ ).

[0031]

Here, the maximum peak temperature ( $T_m$ ) of the endothermic curve measured using the differential scanning calorimeter is obtained from an endothermic curve given when about 5 mg of sample packed in an aluminum pan is heated to  $200^{\circ}\text{C}$  at a heating rate of  $10^{\circ}\text{C}/\text{min}$ , and after the sample is maintained at  $200^{\circ}\text{C}$  for 5 minutes, it is cooled to room temperature at a rate of  $20^{\circ}\text{C}/\text{min}$ , followed by heating at a heating rate of  $10^{\circ}\text{C}/\text{min}$ . This measurement uses an instrument Model DSC-7 (Perkin-Elmer).

[0032]

Such ethylene- $\alpha$ -olefin copolymers having the relationship between the weight fraction ( $W$ ) of the  $n$ -decane-soluble content and the density ( $d$ ) and the relationship between the maximum peak temperature ( $T_m$ ) of the endothermic curve determined using a differential scanning calorimeter (DSC) and density ( $d$ ), as described above, may safely be said to have narrow compositional distributions.

[0033]

Further, the ethylene- $\alpha$ -olefin copolymer (A) employed in the present invention satisfies the following relationship:

$$B_1 \geq B_2$$

wherein  $B_1$  and  $B_2$  are an average branch number of the higher molecular weight part and that of the lower molecular weight part, as determined by GPC-IR respectively.

[0034]

It should be noted here that the average branch number ( $B_1$ ) of the higher molecular weight part as determined by GPC-IR means as follows: Fractions within a cumulative weight fraction range of 15 to 85 % (i.e., excluding the lower 15 % of the low molecular weight region and the higher 15 % of the high molecular weight region) of the polymer eluate fractions obtained by GPC depending on the molecular weight are divided into two parts at the peak molecular weight of the GPC elution curve as a boundary. Branch number values of the fractions are determined respectively, and an average of the thus measured values is taken for each part.  $B_1$  means the average value of the higher molecular weight part, whereas  $B_2$  means that of the lower molecular weight part.

[0035]

The values  $B_1$  and  $B_2$  are determined under the following conditions:

Measuring instrument: Model 1760X (Perkin-Elmer)

Column: TSK gel, GMH-HT (Toso Co. Ltd.)  
(7.5 mm I.D. x 600 mm) x 1

Eluent: o-Dichlorobenzene (ODCB) containing 0.05 % of MP-J (extra pure grade, Wako Pure Chemical Ind., Ltd.)

Column temperature: 140°C  
Sample concentration: 0.1 % (weight/volume)  
Injection volume: 100 µl  
Detector: MCT  
Resolution: 8 cm<sup>-1</sup>

Since the ethylene- $\alpha$ -olefin copolymers (A) having B<sub>1</sub> and B<sub>2</sub> values of the above relationship are of narrow compositional distributions and do not contain large amounts of low polymers, they are less tacky. Accordingly, use of such ethylene- $\alpha$ -olefin copolymers (A) give agricultural films having excellent dust proofness.

[0036]

Further, since the films containing such ethylene- $\alpha$ -olefin copolymers (A) undergo least deterioration of light transmittance with time, these films if used as agricultural films stand long uses.

[0037]

The above-mentioned ethylene- $\alpha$ -olefin copolymers (A) can be produced by copolymerizing ethylene with an  $\alpha$ -olefin having 4 to 12 carbon atoms in the presence of a single-site catalyst, for example, the so-called metallocene catalyst for olefin polymerization including metallocene catalyst components described, for example, in Japanese Unexamined Patent publication Nos. Hei 6-9724, 6-136195, 6-136196 and 6-207057 so that the resultant copolymer comes to have a density of 0.905 to 0.935 g/cm<sup>3</sup>.

[0038]

Such a metallocene catalyst is generally formed from a metallocene catalyst component (a) containing a compound of a transition metal belonging to Group IVB of the Periodic

Table, which compound has at least one ligand having a cyclopentadienyl skeleton; an organoaluminumoxy compound catalyst component (b); a particulate carrier (c); and if desired, an organoaluminum compound catalyst component (d) and an ionized ionic compound catalyst component (e).

[0039]

Metallocene catalyst components (a) suitably used in the present invention includes compounds of transition metals belonging to Group IVB of the Periodic Table each containing at least one ligand having a cyclopentadienyl skeleton. Such transition metal compounds include, for example, those represented by the following general formula [I]:

[0040]



[I].

In the above formula, x represents a valence of transition metal atom M; M represents a transition metal atom selected from those belonging to Group IVB of the Periodic Table, and is typically zirconium, titanium or hafnium. Of these metals, preferred is zirconium.

[0041]

$L^1$  represents ligands ligated to the transition metal atom M, and at least one of them is a ligand having a cyclopentadienyl skeleton. Specific examples of such ligands  $L^1$  having the cyclopentadienyl skeleton ligated to the transition metal atom M include, for example, alkyl-substituted cyclopentadienyl groups, such as a cyclopentadienyl group, a methylcyclopentadienyl group, a dimethylcyclopentadienyl group, a trimethylcyclopentadienyl group, a tetramethylcyclopentadienyl group, a pentamethylcyclopentadienyl group, a

methylethylcyclopentadienyl group and a hexylcyclopentadienyl group; an indenyl group; a 4,5,6,7-tetrahydroindenyl group; a fluorenyl group. These groups may be substituted with a halogen atom, a trialkylsilyl group or the like.

[0042]

When the compound represented by the above general formula [I] has two or more groups having the cyclopentadienyl skeleton, two of them having the cyclopentadienyl skeleton may be bonded to each other through an alkylene group, such as ethylene and propylene; a substituted alkylene group, such as isopropylidenediphenylmethylene; a silylene group; or a substituted silylene group, such as a dimethylsilylene group, a diphenylsilylene group and a methylphenylsilylene group.

[0043]

The ligands  $L^1$  other than those having the cyclopentadienyl skeleton include a hydrocarbon group having 1 to 12 carbon atoms; an alkoxyl group, such as a methoxy group; an aryloxy group, such as a phenoxy group; a trialkylsilyl group, such as a trimethylsilyl group and a triphenylsilyl group;  $SO_3R$  (wherein R represents a hydrocarbon group having 1 to 8 carbon atoms, which may have a substituent, such as a halogen); a halogen atom; or a hydrogen atom.

[0044]

Examples of hydrocarbon groups having 1 to 12 carbon atoms include alkyl groups, such as a methyl group; cycloalkyl groups, such as a cyclopentyl group; aryl groups, such as a phenyl group; aralkyl groups, such as a benzyl

group and the like.

[0045]

Specific examples of ligands represented by  $\text{SO}_3\text{R}$  include, for example, a p-toluenesulfonato group, a methanesulfonato group and a trifluoromethanesulfonato group. As the organoaluminumoxy compound catalyst component (b), aluminoxane is preferably used. Typically, there are used methylaluminoxane, ethylaluminoxane, methylethylaluminoxane and the like, each having generally about 3 to about 50 repeating units represented by  $-\text{Al}(\text{R})\text{O}-$  (wherein R represents an alkyl group).

[0046]

Such an aluminoxane can be prepared by the conventional preparation method. The particulate carrier (c) used in the preparation of an olefin polymerization catalyst is an inorganic or organic compound, and generally has a particle diameter of about 10 to 300  $\mu\text{m}$ , preferably a granular or particulate solid of 20 to 200  $\mu\text{m}$ .

[0047]

The inorganic carrier, includes preferably porous oxides typically, for example,  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{MgO}$ ,  $\text{ZrO}_2$ ,  $\text{TiO}_2$ ,  $\text{B}_2\text{O}_3$ ,  $\text{CaO}$ ,  $\text{ZnO}$ ,  $\text{BaO}$  and  $\text{SnO}_2$  or mixtures of them. The above-mentioned inorganic oxides may each contain a small amount of carbonate such as  $\text{Na}_2\text{CO}_3$ , a sulfate such as  $\text{Al}_2(\text{SO}_4)_3$ , a nitrate such as  $\text{KNO}_3$ , or an oxide such as  $\text{LiO}_2$ .

[0048]

While properties of such carriers vary depending on the type and the preparation method, and it is desired that the carrier suitably used in the present invention has a

specific surface area of 50 to 1,000 m<sup>2</sup>/g, preferably 100 to 700 m<sup>2</sup>/g, and a pore volume of 0.3 to 2.5 cm<sup>3</sup>/g.

[0049]

This carrier is used, if desired, after calcination at 100 to 1,000°C, preferably 150 to 700°C. Further, examples of organic compounds used as the particulate carrier include (co)polymers formed to contain as a major component an  $\alpha$ -olefin having 2 to 14 carbon atoms, such as ethylene and 4-methyl-1-pentene, and (co)polymers formed to contain as a major component vinylcyclohexane or styrene.

[0050]

Specific examples of the organoaluminum compound catalyst component (d) used if desired in the preparation of an olefin polymerization catalyst include a trialkylaluminum such as trimethylaluminum; an alkenylaluminum such as isoprenylaluminum; a dialkylaluminum halide such as dimethylaluminum chloride; an alkylaluminum sesquihalide such as methylaluminum sesquichloride; an alkylaluminum dihalide such as methylaluminum dichloride; an alkylaluminum hydride such as diethylaluminum hydride.

[0051]

The ionized ionic compound catalyst component (e) includes, for example, Lewis acids such as triphenylboron, MgCl<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>; ionic compounds such as triphenylcarbenium tetrakis(pentafluorophenyl)borate; and carborane compounds such as dodecarborane, bis-n-butylammonium (1-carbedodeca)borate and the like, as described in U.S. Patent No. 5,321,106.

[0052]

The ethylene- $\alpha$ -olefin copolymer (A) used in the present invention can be obtained by copolymerizing ethylene with an  $\alpha$ -olefin having 4 to 12 carbon atoms in the presence of such an olefin polymerization catalyst as described above containing the metallocene catalyst component (a), organoaluminumoxy compound catalyst component (b), particulate carrier (c), and if desired, organoaluminum compound catalyst component (d) and ionized ionic compound catalyst component (e) in a gas phase in the form or liquid phase in the form of slurry or solution under various conditions.

[0053]

In the slurry polymerization method or solution polymerization method, an inert hydrocarbon or the olefin per se may be used as a solvent. In carrying out the polymerization, it is desired that the above-mentioned metallocene catalyst for olefin polymerization is used generally in an amount of  $10^{-8}$  to  $10^{-3}$  gram atom/liter, preferably  $10^{-7}$  to  $10^{-4}$  gram atom/liter, in terms of the concentration of the transition metal atom in the polymerization reaction system.

[0054]

Meanwhile, in the polymerization, an organoaluminumoxy compound catalyst component (b) and/or an organoaluminum compound catalyst component (d) which are not carried on a carrier may be further used in addition to the organoaluminumoxy compound catalyst component (b) and organoaluminum compound catalyst component (d) which are carried on a carrier. In such a case, the atomic ratio [Al/M] of the aluminum atom (Al) derived from the

organoaluminumoxy compound catalyst component (b) and/or organoaluminum compound catalyst component (d) which are not carried on a carrier to the transition metal atom (M) derived from the metallocene catalyst component (a) is in the range of from 5 to 300, preferably 10 to 200, more preferably from 15 to 150.

[0055]

The polymerization temperature in the slurry polymerization method is generally in the range of from -50 to 100°C, preferably from 0 to 90°C, and the polymerization temperature in the solution polymerization method is generally in the range of from -50 to 500°C, preferably from 0 to 400°C. Meanwhile, the polymerization temperature in the gas phase polymerization method is generally in the range of from 0 to 120°C, preferably from 20 to 100°C.

[0056]

The polymerization is conducted generally under atmospheric pressure or under pressurizing conditions of 100 kg/cm<sup>2</sup>, preferably 2 to 50 kg/cm<sup>2</sup> batchwise, semi-continuously or continuously.

[0057]

In the present invention, in the preparation of the above-mentioned ethylene- $\alpha$ -olefin copolymer (A), if desired, there can be employed, for example, (1) a multi-stage polymerization, (2) a multi-stage polymerization in a liquid phase and a gas phase, or (3) a method in which polymerization is conducted in a gas phase after prepolymerization in a liquid phase. In the present invention, the multi-stage polymerization (1) is preferred.

[0058]

The multi-stage polymerization method can be exemplified as follows:

A multi-stage olefin polymerization method including [1] a step of copolymerizing ethylene with an  $\alpha$ -olefin having 4 to 12 carbon atoms in the presence of a metallocene catalyst containing a metallocene catalyst component of at least one compound selected from those of transition metals belonging to Group IVB of the Periodic Table, which compounds are represented by the above general formula [I] and contain a ligand having the cyclopentadienyl skeleton, and an organoaluminumoxy compound catalyst component, to prepare ethylene- $\alpha$ -olefin copolymer (A-1), and [2] a step of copolymerizing ethylene with an  $\alpha$ -olefin having 4 to 12 carbon atoms in the presence of a metallocene catalyst containing a metallocene catalyst component of at least one compound selected from the compounds of transition metals belonging to Group IVB of the Periodic Table, which compounds are represented by the above general formula [I] and contain a ligand having the cyclopentadienyl skeleton, and an organoaluminumoxy compound catalyst component, in a polymerizer different from that used in the above-mentioned polymerization reaction, to prepare ethylene- $\alpha$ -olefin copolymer (A-2). It is noted that the preparation conditions in step [1] are different from those in step [2]. Such preparation conditions include, for example, the kind and amount of the metallocene catalyst component, the kind and amount of the organoaluminumoxy compound catalyst component and the molar ratio of ethylene to the  $\alpha$ -olefin.

[0059]

The metallocene catalyst used in the above

copolymerization step [1] and/or [2] may be either a catalyst containing in addition to the metallocene catalyst component (a) and organoaluminumoxy compound catalyst component (b), an organoaluminum compound catalyst component (d) or a solid catalyst having the metallocene catalyst component (a) and organoaluminumoxy compound catalyst component (b) carried on a particulate carrier (c). In addition, these metallocene catalysts may be a prepolymerization catalyst of a solid catalyst component, having the metallocene catalyst component (a) and organoaluminumoxy compound catalyst component (b) carried on a particulate carrier (c), prepolymerized with an olefin. Further, these metallocene catalysts may be those each having the above-mentioned solid catalyst (solid catalyst component) and the organoaluminum compound catalyst component (d) or those each having the above-mentioned prepolymerization catalyst (prepolymerization catalyst compound) and the organoaluminum compound catalyst component (d).

[0060]

In the multi-stage polymerization method, a plurality of polymerizers connected to one another in series are used. The ethylene- $\alpha$ -olefin copolymer (A-1) is first prepared in one polymerizer and then transferred therefrom into another polymerizer and the ethylene- $\alpha$ -olefin copolymer (A-2) can be prepared in the presence of ethylene- $\alpha$ -olefin copolymer (A-1).

[0061]

Alternatively, a plurality of polymerizers are connected to one another in parallel, and ethylene- $\alpha$ -olefin copolymers (A-1) and (A-2) are prepared in the polymerizers

respectively, and then, the thus produced copolymers can be blended.

[0062]

In the present invention, ethylene- $\alpha$ -olefin copolymer (A) is used in a proportion of 60 to 100 parts by weight, preferably 65 to 99 parts by weight, more preferably 70 to 95 parts by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (A) and high-pressure-processed low-density polyethylene (B).

[0063]

[High-pressure-processed low-density polyethylene (B)]

In the present invention, for improving the molding property of the ethylene- $\alpha$ -olefin copolymer (A), the high-pressure-processed low-density polyethylene (B) can be used in admixture with the ethylene- $\alpha$ -olefin copolymer (A).

[0064]

It is preferred that the high-pressure-processed low-density polyethylene (B) constituting the ethylene copolymer composition (1) used in the present invention has a melt flow rate (MFR; ASTM D 1238, measured at 190°C under a load of 2.16 kg) of 0.1 to 100 g/10 minutes, a density of 0.915 to 0.935 g/cm<sup>3</sup>, and a swell ratio of 60 % or less.

[0065]

The density of the high-pressure-processed low-density polyethylene (B) is determined according to the same method as used for measuring the density of the ethylene- $\alpha$ -olefin copolymer (A). Meanwhile, the swell ratio is determined as follows.

[0066]

The diameter of the strand obtained in the measurement of melt flow rate at a distance of 5 mm from the tip thereof is measured by a micrometer as a diameter (mm) of the sample. Then, the swell ratio is calculated according to the following formula:

[0067]

$$\text{Swell ratio (\%)} = [(L_1 / L_0) - 1] \times 100$$

wherein  $L_1$  represents a sample diameter (mm),  
and  $L_0$  represents an orifice diameter (= 2.0955 mm).

The above-mentioned high-pressure-processed low-density polyethylene (B) can be produced by the conventionally known high pressure process.

[0068]

In the present invention, high-pressure-processed low-density polyethylene (B) is used in a proportion of 0 to 40 parts by weight, preferably 1 to 35 parts by weight, more preferably 5 to 30 parts by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (A) and high-pressure-processed low-density polyethylene (B). When the high-pressure-processed low-density polyethylene (B) is mixed with the ethylene- $\alpha$ -olefin copolymer (A) in the above-mentioned proportion, it is possible to improve molding properties of ethylene- $\alpha$ -olefin copolymer (A) and to improve clarity of the molded film.

[0069]

[Anti-fogging agent (C)]

The anti-fogging agent (C) constituting the ethylene copolymer composition (1) used in the present invention is

a mixture of two or more kinds of nonionic surfactants, and contains in the mixture a polyoxyethylene-containing component in an amount of 15 to 50 % by weight, preferably 20 to 45 % by weight, more preferably 25 to 40 % by weight. It is noted that diethanolalkylamines are excluded from the nonionic surfactant used in the present invention. When the polyoxyethylene-containing component is used in the above-mentioned proportion, the resultant composition has excellent molding properties and the film obtained from the composition has excellent anti-fogging properties and excellent sustained anti-fogging properties.

[0070]

The above polyoxyethylene-containing component has a polyoxyethylene group represented by  $(-\text{CH}_2\text{CH}_2\text{O}-)_n\text{H}$  wherein  $n$  is 2 to 10, and the HLB (hydrophilic-lipophilic balance) value thereof, which is an index of the balance between hydrophilicity and lipophilicity, is suitably 5 to 12.

[0071]

The mixture of two or more kinds of nonionic surfactants suitably used in the present invention as anti-fogging agent (C) contains (i) at least one kind of fatty acid ester selected from the group consisting of glycerol fatty acid esters, polyglycerol fatty acid esters and sorbitan fatty acid esters, and (ii) at least one kind of polyoxyethylene-containing component selected from the group consisting of ethylene oxide addition products of glycerol fatty acid esters (polyoxyethylene glycerol fatty acid ester), ethylene oxide addition products of polyglycerol fatty acid esters (polyoxyethylene polyglycerol fatty acid ester), and ethylene oxide addition products of sorbitan fatty acid esters (polyoxyethylene sorbitan fatty acid ester).

[0072]

Specific examples of the above-mentioned glycerol fatty acid esters include higher fatty acid esters, such as glycerol monomyristate, glycerol dimyristate, glycerol monopalmitate and glycerol dipalmitate, glycerol monostearate, glycerol distearate, glycerol monooleate, glycerol dioleate, glycerol monolaurate, glycerol dilaurate, glycerol monobehenate and glycerol dibehenate, and mixtures of two or more kinds of such esters. Especially preferred of all is glycerol monostearate.

[0073]

Specific examples of the above-mentioned polyglycerol fatty acid esters include higher fatty acid esters, such as diglycerol fatty acid esters, e.g., diglycerol monomyristate, diglycerol dimyristate, diglycerol monopalmitate, diglycerol dipalmitate, diglycerol monostearate, diglycerol distearate, diglycerol monooleate, diglycerol dioleate, diglycerol monolaurate, diglycerol dilaurate, diglycerol monobehenate and diglycerol dibehenate; triglycerol fatty acid esters, e.g., triglycerol monomyristate, triglycerol dimyristate, triglycerol monopalmitate, triglycerol dipalmitate, triglycerol monostearate, triglycerol distearate, triglycerol monooleate, triglycerol dioleate, triglycerol monolaurate, triglycerol dilaurate, triglycerol monobehenate and triglycerol dibehenate; and mixtures of two or more kinds of such esters. Of these, especially preferred are diglycerol monostearate and diglycerol distearate.

[0074]

The above-mentioned sorbitan fatty acid ester is an

ester of a sorbitan mixture and a higher fatty acid. Sorbitan is generally industrially produced as a mixture of 1,4-sorbitan, 3,6-sorbitan or 1,5-sorbitan, which is a one-molecule dehydrate of sorbitol, and 1,4,3,6-sorbid which is a two-molecule dehydrate of sorbitol. The higher fatty acid is exemplified by lauric acid, palmitic acid, stearic acid, oleic acid, myristic acid and behenic acid.

[0075]

Specific examples of sorbitan fatty acid esters include higher fatty acid esters, such as sorbitan monolaurate, sorbitan dilaurate, sorbitan trilaurate, sorbitan monopalmitate, sorbitan dipalmitate, sorbitan tripalmitate, sorbitan monostearate, sorbitan distearate, sorbitan tristearate, sorbitan monooleate, sorbitan dioleate, sorbitan trioleate, sorbitan monomyristate, sorbitan dimyristate, sorbitan trimyristate, sorbitan monobehenate, sorbitan dibehenate and sorbitan tribehenate, and mixtures of two or more kinds of such esters. Especially preferred of all are sorbitan monostearate, sorbitan distearate and a mixture thereof.

[0076]

The above-mentioned ethylene oxide addition products of glycerol fatty acid esters include specifically, for example, the ethylene oxide addition products of glycerol fatty acid esters mentioned above as specific examples, e.g., dioxyethylene glycerol myristate (including mono-, di- and triesters), dioxyethylene glycerol palmitate (including mono-, di- and triesters), dioxyethylene glycerol stearate (including mono-, di- and triesters), trioxyethylene glycerol myristate (including mono-, di- and triesters), trioxyethylene glycerol palmitate (including mono-, di- and triesters), trioxyethylene glycerol stearate

(including mono-, di- and triesters), tetraoxyethylene glycerol myristate (including mono-, di- and triesters), tetraoxyethylene glycerol palmitate (including mono-, di- and triesters), tetraoxyethylene glycerol stearate (including mono-, di- and triesters), pentaoxyethylene glycerol myristate (including mono-, di- and triesters), pentaoxyethylene glycerol palmitate (including mono-, di- and triesters), pentaoxyethylene glycerol stearate (including mono-, di- and triesters), hexaoxyethylene glycerol myristate (including mono-, di- and triesters), hexaoxyethylene glycerol palmitate (including mono-, di- and triesters), hexaoxyethylene glycerol stearate (including mono-, di- and triesters), heptaoxyethylene glycerol myristate (including mono-, di- and triesters), heptaoxyethylene glycerol palmitate (including mono-, di- and triesters), heptaoxyethylene glycerol stearate (including mono-, di- and triesters), octaoxyethylene glycerol myristate (including mono-, di- and triesters), octaoxyethylene glycerol palmitate (including mono-, di- and triesters), octaoxyethylene glycerol stearate (including mono-, di- and triesters), nonaoxyethylene glycerol myristate (including mono-, di- and triesters), nonaoxyethylene glycerol palmitate (including mono-, di- and triesters), nonaoxyethylene glycerol stearate (including mono-, di- and triesters), decaoxyethylene glycerol myristate (including mono-, di- and triesters), decaoxyethylene glycerol palmitate (including mono-, di- and triesters) and decaoxyethylene glycerol stearate (including mono-, di- and triesters).

[0077]

The above-mentioned ethylene oxide addition products of polyglycerol fatty acid esters include specifically, for example, the ethylene oxide addition products of

polyglycerol fatty acid esters mentioned above as specific examples, e.g., dioxyethylene diglycerol myristate (including mono-, di-, tri- and tetraesters), dioxyethylene diglycerol palmitate (including mono-, di-, tri- and tetraesters), dioxyethylene diglycerol stearate (including mono-, di-, tri- and tetraesters), dioxyethylene triglycerol myristate (including mono-, di-, tri- and tetraesters), dioxyethylene triglycerol palmitate (including mono-, di-, tri- and tetraesters), dioxyethylene triglycerol stearate (including mono-, di-, tri- and tetraesters), trioxyethylene diglycerol myristate (including mono-, di-, tri- and tetraesters), trioxyethylene diglycerol palmitate (including mono-, di-, tri- and tetraesters), trioxyethylene diglycerol stearate (including mono-, di-, tri- and tetraesters), tetraoxyethylene diglycerol myristate (including mono-, di-, tri- and tetraesters), tetraoxyethylene diglycerol palmitate (including mono-, di-, tri- and tetraesters), tetraoxyethylene diglycerol stearate (including mono-, di-, tri- and tetraesters), pentaoxyethylene diglycerol myristate (including mono-, di-, tri- and tetraesters), pentaoxyethylene diglycerol palmitate (including mono-, di-, tri- and tetraesters), pentaoxyethylene diglycerol stearate (including mono-, di-, tri- and tetraesters), hexaoxyethylene diglycerol myristate (including mono-, di-, tri- and tetraesters), hexaoxyethylene diglycerol palmitate (including mono-, di-, tri- and tetraesters), hexaoxyethylene diglycerol stearate (including mono-, di-, tri- and tetraesters), heptaoxyethylene diglycerol myristate (including mono-, di-, tri- and tetraesters), heptaoxyethylene diglycerol palmitate (including mono-, di-, tri- and tetraesters), heptaoxyethylene diglycerol stearate (including mono-, di-, tri- and tetraesters), octaoxyethylene diglycerol myristate (including mono-, di-,

tri- and tetraesters), octaoxyethylene diglycerol palmitate (including mono-, di-, tri- and tetraesters), octaoxyethylene diglycerol stearate (including mono-, di-, tri- and tetraesters), nonaoxyethylene diglycerol myristate (including mono-, di-, tri- and tetraesters), nonaoxyethylene diglycerol palmitate (including mono-, di-, tri- and tetraesters), nonaoxyethylene diglycerol stearate (including mono-, di-, tri- and tetraesters), decaoxyethylene diglycerol myristate (including mono-, di-, tri- and tetraesters), decaoxyethylene diglycerol palmitate (including mono-, di-, tri- and tetraesters) and decaoxyethylene diglycerol stearate (including mono-, di-, tri- and tetraesters).

[0078]

As the above-mentioned ethylene oxide addition products of sorbitan fatty acid esters include specifically, for example, the ethylene oxide addition products of sorbitan fatty acid esters mentioned above as specific examples, e.g., dioxyethylene sorbitan laurate (including mono-, di-, tri- and tetraesters), dioxyethylene sorbitan palmitate (including mono-, di-, tri- and tetraesters), dioxyethylene sorbitan stearate (including mono-, di-, tri- and tetraesters), dioxyethylene sorbitan oleate (including mono-, di-, tri- and tetraesters), trioxyethylene sorbitan laurate (including mono-, di-, tri- and tetraesters), trioxyethylene sorbitan palmitate (including mono-, di-, tri- and tetraesters), trioxyethylene sorbitan stearate (including mono-, di-, tri- and tetraesters), trioxyethylene sorbitan oleate (including mono-, di-, tri- and tetraesters), tetraoxyethylene sorbitan laurate (including mono-, di-, tri- and tetraesters), tetraoxyethylene sorbitan palmitate (including mono-, di-, tri- and tetraesters), tetraoxyethylene sorbitan stearate

(including mono-, di-, tri- and tetraesters),  
 tetraoxyethylene sorbitan oleate (including mono-, di-,  
 tri- and tetraesters), pentaoxyethylene sorbitan laurate  
 (including mono-, di-, tri- and tetraesters),  
 pentaoxyethylene sorbitan palmitate (including mono-, di-,  
 tri- and tetraesters), pentaoxyethylene sorbitan stearate  
 (including mono-, di-, tri- and tetraesters),  
 pentaoxyethylene sorbitan oleate (including mono-, di-,  
 tri- and tetraesters), hexaoxyethylene sorbitan laurate  
 (including mono-, di-, tri- and tetraesters),  
 hexaoxyethylene sorbitan palmitate (including mono-, di-,  
 tri- and tetraesters), hexaoxyethylene sorbitan stearate  
 (including mono-, di-, tri- and tetraesters),  
 hexaoxyethylene sorbitan oleate (including mono-, di-, tri-  
 and tetraesters), heptaoxyethylene sorbitan laurate  
 (including mono-, di-, tri- and tetraesters),  
 heptaoxyethylene sorbitan palmitate (including mono-, di-,  
 tri- and tetraesters), heptaoxyethylene sorbitan stearate  
 (including mono-, di-, tri- and tetraesters),  
 heptaoxyethylene sorbitan oleate (including mono-, di-,  
 tri- and tetraesters), octaoxyethylene sorbitan laurate  
 (including mono-, di-, tri- and tetraesters),  
 octaoxyethylene sorbitan palmitate (including mono-, di-,  
 tri- and tetraesters), octaoxyethylene sorbitan stearate  
 (including mono-, di-, tri- and tetraesters),  
 octaoxyethylene sorbitan oleate (including mono-, di-, tri-  
 and tetraesters), nonaoxyethylene sorbitan laurate  
 (including mono-, di-, tri- and tetraesters),  
 nonaoxyethylene sorbitan palmitate (including mono-, di-,  
 tri- and tetraesters), nonaoxyethylene sorbitan stearate  
 (including mono-, di-, tri- and tetraesters),  
 nonaoxyethylene sorbitan oleate (including mono-, di-, tri-  
 and tetraesters), decaoxyethylene sorbitan laurate  
 (including mono-, di-, tri- and tetraesters),

decaoxyethylene sorbitan palmitate (including mono-, di-, tri- and tetraesters), decaoxyethylene sorbitan stearate (including mono-, di-, tri- and tetraesters) and decaoxyethylene sorbitan oleate (including mono-, di-, tri- and tetraesters).

[0079]

These polyoxyethylene-containing components each have a polyoxyethylene group represented by  $(-\text{CH}_2\text{CH}_2\text{O}-)_n\text{H}$  as mentioned above, and it is preferred that  $n$  in this formula is 2 to 10, and the HLB value is 5 to 12.

[0080]

It is desired that the glycerol fatty acid ester is contained in a proportion of 0 to 60 parts by weight, preferably 10 to 50 parts by weight, while the polyglycerol fatty acid ester is contained in a proportion of 0 to 80 parts by weight, preferably 20 to 60 parts by weight, and the sorbitan fatty acid ester is contained in a proportion of 0 to 50 parts by weight, preferably 10 to 40 parts by weight, per 100 parts by weight of the total amount of the glycerol fatty acid ester, the polyglycerol fatty acid ester and the sorbitan fatty acid ester. The total weight of the glycerol fatty acid ester, the polyglycerol fatty acid ester and the sorbitan fatty acid ester is 50 to 85 % by weight, preferably 55 to 80 % by weight, more preferably 60 to 75 % by weight, based on the total amount of anti-fogging agent (C).

[0081]

Meanwhile, it is desired that the polyoxyethylene glycerol fatty acid ester is contained in a proportion of 0 to 60 parts by weight, preferably 10 to 50 parts by weight, while the polyoxyethylene polyglycerol fatty acid ester is

contained in a proportion of 0 to 80 parts by weight, preferably 20 to 60 parts by weight, and the polyoxyethylene sorbitan fatty acid ester is contained in a proportion of 0 to 50 parts by weight, preferably 10 to 40 parts by weight, per 100 parts by weight of the total amount of the polyoxyethylene glycerol fatty acid ester, the polyoxyethylene polyglycerol fatty acid ester and the polyoxyethylene sorbitan fatty acid ester. The total amount of the polyoxyethylene glycerol fatty acid ester, the polyoxyethylene polyglycerol fatty acid ester and the polyoxyethylene sorbitan fatty acid ester which are the polyoxyethylene-containing components is 15 to 50 % by weight, preferably 20 to 45 % by weight, more preferably 25 to 40 % by weight, based on the total amount of anti-fogging agent (C).

[0082]

When different anti-fogging agents are used in combination in the proportion within the range as mentioned above, the anti-fogging effect is maintained from the initial stage over a long term, favorably. Anti-fogging agents (C) suitably used in the present invention are, for example, as follows.

- (1) Anti-fogging agent containing glycerol monostearate, and polyoxyethylene glycerol monostearate and/or polyoxyethylene glycerol distearate,
- (2) Anti-fogging agent containing diglycerol monostearate and/or diglycerol distearate, and polyoxyethylene glycerol monostearate and/or polyoxyethylene glycerol distearate,
- (3) Anti-fogging agent containing sorbitan monostearate and/or sorbitan distearate, and polyoxyethylene glycerol monostearate and/or polyoxyethylene glycerol distearate,
- (4) Anti-fogging agent containing glycerol monostearate, and diglycerol monostearate and/or diglycerol distearate,

and polyoxyethylene glycerol monostearate and/or  
polyoxyethylene glycerol distearate,

(5) Anti-fogging agent containing glycerol monostearate,  
and sorbitan monostearate and/or sorbitan distearate, and  
polyoxyethylene glycerol monostearate and/or  
polyoxyethylene glycerol distearate,

(6) Anti-fogging agent containing diglycerol monostearate  
and/or diglycerol distearate, and sorbitan monostearate  
and/or sorbitan distearate, and polyoxyethylene glycerol  
monostearate and/or polyoxyethylene glycerol distearate,

(7) Anti-fogging agent containing glycerol monostearate,  
and diglycerol monostearate and/or diglycerol distearate,  
and sorbitan monostearate and/or sorbitan distearate, and  
polyoxyethylene glycerol monostearate and/or  
polyoxyethylene glycerol distearate,

(8) Anti-fogging agent containing glycerol monostearate,  
and polyoxyethylene diglycerol monostearate and/or  
polyoxyethylene diglycerol distearate,

(9) Anti-fogging agent containing diglycerol monostearate  
and/or diglycerol distearate, and polyoxyethylene  
diglycerol monostearate and/or polyoxyethylene diglycerol  
distearate,

(10) Anti-fogging agent containing sorbitan monostearate  
and/or sorbitan distearate, and polyoxyethylene diglycerol  
monostearate and/or polyoxyethylene diglycerol distearate,

(11) Anti-fogging agent containing glycerol monostearate,  
and diglycerol monostearate and/or diglycerol distearate,  
and polyoxyethylene diglycerol monostearate and/or  
polyoxyethylene diglycerol distearate,

(12) Anti-fogging agent containing glycerol monostearate,  
and sorbitan monostearate and/or sorbitan distearate, and  
polyoxyethylene diglycerol monostearate and/or  
polyoxyethylene diglycerol distearate,

(13) Anti-fogging agent containing diglycerol monostearate

and/or diglycerol distearate, and sorbitan monostearate and/or sorbitan distearate, and polyoxyethylene diglycerol monostearate and/or polyoxyethylene diglycerol distearate, (14) Anti-fogging agent containing glycerol monostearate, and diglycerol monostearate and/or diglycerol distearate, and sorbitan monostearate and/or sorbitan distearate, and polyoxyethylene diglycerol monostearate and/or

polyoxyethylene diglycerol distearate,

(15) Anti-fogging agent containing glycerol monostearate, and polyoxyethylene sorbitan monostearate and/or polyoxyethylene sorbitan distearate,

(16) Anti-fogging agent containing diglycerol monostearate and/or diglycerol distearate, and polyoxyethylene sorbitan monostearate and/or polyoxyethylene sorbitan distearate,

(17) Anti-fogging agent containing glycerol monostearate, and diglycerol monostearate and/or diglycerol distearate, and polyoxyethylene sorbitan monostearate and/or polyoxyethylene sorbitan distearate,

(18) Anti-fogging agent containing glycerol monostearate, and sorbitan monostearate and/or sorbitan distearate, and polyoxyethylene sorbitan monostearate and/or polyoxyethylene sorbitan distearate,

(19) Anti-fogging agent containing diglycerol monostearate and/or diglycerol distearate, and sorbitan monostearate and/or sorbitan distearate, and polyoxyethylene sorbitan monostearate and/or polyoxyethylene sorbitan distearate and

(20) Anti-fogging agent containing glycerol monostearate, and diglycerol monostearate and/or diglycerol distearate, and sorbitan monostearate and/or sorbitan distearate, and polyoxyethylene sorbitan monostearate and/or polyoxyethylene sorbitan distearate.

[0083]

The anti-fogging agent (C) is used in a proportion of

0.05 to 5 parts by weight, preferably 0.1 to 4 parts by weight, more preferably 0.5 to 3 parts by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (A) and the high-pressure-processed low-density polyethylene (B).

[0084]

Further, for the purpose of enhancing maintenance of the anti-fogging agent (C) in the film and improving further sustained anti-fogging properties, a modified wax can be added in a proportion of 0.1 to 5 parts by weight, preferably 0.5 to 4 parts by weight, more preferably 1 to 3 parts by weight, per 100 parts by weight of the total amount of ethylene- $\alpha$ -olefin copolymer (A) and the high-pressure-processed low-density polyethylene (B). Specific examples of such modified wax include maleic acid modified polyethylene wax, acrylic acid modified polyethylene wax, oxidized polyethylene wax and the like.

[0085]

The modified wax generally has an acid value of 1 to 60 KOH mg/g and a molecular weight (Mv) of 1,000 to 5,000.

[Nonionic fluorine-containing surfactant (D)]

The nonionic fluorine-containing surfactant (D) used in the present invention has a fluorine content of 25 to 65 % by weight, and generally has a surface tension of 42 dyne/cm or less at 25°C as a 0.001 % aqueous solution. In the present invention, an organic fluorine compound having a perfluoroalkyl group is especially preferred.

[0086]

Such organic fluorine compounds include, for example,

alkylene oxide addition products of perfluoroalkyl alcohols, such as perfluoroalkylethylene oxide addition products and perfluoroalkylpropylene oxide addition products; oligomers having a perfluoroalkyl group, such as perfluoroalkyl acrylate oligomers and perfluoroalkyl methacrylate oligomers.

[0087]

The above-mentioned perfluoroalkylethylene oxide addition product is commercially available, for example, from DAIKIN INDUSTRIES, Ltd. under the trade name of DS-403, and from Asahi Glass Co., Ltd. under the trade name of KC-40.

[0088]

Meanwhile, the above-mentioned oligomer having a perfluoroalkyl group is commercially available, for example, from Asahi Glass Co., Ltd. under the trade name of S-393. Further, a mixture product of a perfluoroalkylethylene oxide addition product and a perfluoroalkyl-containing oligomer is commercially available, for example, from Asahi Glass Co., Ltd. under the trade name of KC-14.

[0089]

The nonionic fluorine-containing surfactant (D) is used in a proportion of 0.01 to 1 part by weight, preferably 0.05 to 0.3 part by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (A) and the high-pressure-processed low-density polyethylene (B). Conventionally, films incorporated with fluorine compounds come to have reduced anti-fogging properties, as compared with those having no anti-fogging agent incorporated thereto. However, the present inventors have found that anti-fogging properties of the films can be

improved unexpectedly by using the nonionic fluorine-containing surfactant (D) in the above-mentioned proportion in the raw materials for the film of the present invention and the composition thereof and the films show excellent sustained anti-fogging properties (long-term anti-fogging properties).

[0090]

[Other components]

To the ethylene copolymer composition (1) may be added, if desired, additives, such as the conventionally known weathering agents, antioxidants, inorganic compounds, antistatic agents and thermal stabilizers, so long as the intended objective of the present invention is not sacrificed.

[0091]

The weathering agent is roughly classified into ultraviolet absorbers and light stabilizers, and they can be used independently or in combination, to show a remarkable effect of improving weathering stability.

[0092]

As the light stabilizer, the conventionally known light stabilizers can be used, hindered amine light stabilizers (HALS) are preferably used among others.

[0093]

Hindered amine stabilizers employable specifically include, for example, the following compounds:

(1) bis(2,2,6,6-tetramethyl-4-piperidyl) sebacate, (2) dimethyl succinate-1-(2-hydroxyethyl)-4-hydroxy-2,2,6,6-tetramethylpiperidine condensation product, (3) tetrakis(2,2,6,6-tetramethyl-4-piperidyl)-1,2,3,4-

butanetetracarboxylate, (4) 2,2,6,6-tetramethyl-4-piperidiny1 benzoate, (5) bis(1,2,6,6-tetramethyl-4-piperidiny1)-2-(3,5-di-t-butyl-4-hydroxybenzyl)-2-n-butyl malonate, (6) bis(N-methyl-2,2,6,6-tetramethyl-4-piperidiny1) sebacate, (7) 1,1'-(1,2-ethanediyl)bis(3,3,5,5-tetramethylpiperazinone), (8) (mixed 2,2,6,6-tetramethyl-4-piperidyl/tridecyl)-1,2,3,4-butanetetracarboxylate, (9) (mixed 1,2,2,6,6-pentamethyl-4-piperidyl/tridecyl)-1,2,3,4-butanetetracarboxylate, (10) mixed {2,2,6,6-tetramethyl-4-piperidyl/ $\beta$ , $\beta$ , $\beta'$ , $\beta'$ -tetramethyl-3-9-[2,4,8,10-tetraoxaspiro(5,5)undecane]diethyl}-1,2,3,4-butanetetracarboxylate, (11) mixed {1,2,2,6,6-pentamethyl-4-piperidyl/ $\beta$ , $\beta$ , $\beta'$ , $\beta'$ -tetramethyl-3-9-[2,4,8,10-tetraoxaspiro(5,5)undecane]diethyl}-1,2,3,4-butanetetracarboxylate, (12) N,N'-bis(3-aminopropyl)ethylenediamine-2-4-bis[N-butyl-N-(1,2,2,6,6-pentamethyl-4-piperidyl)amino]-6-chloro-1,3,5-triazine condensation product, (13) condensation product of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)hexamethylenediamine and 1,2-dibromoethane, (14) [N-(2,2,6,6-tetramethyl-4-piperidyl)-2-methyl-2-(2,2,6,6-tetramethyl-4-piperidyl)imino]propionamide, and (15) poly{[6-[(1,1,3,3-tetramethylbutyl)amino]-1,3,5-triazine-2,4-diyl][[(2,2,6,6-tetramethyl-4-piperidyl)imino]hexamethylene[(2,2,6,6-tetramethyl-4-piperidyl)imino]]} (trade name: Kimasorb 944).

[0094]

These hindered amine light stabilizers can be used independently or in combination of two or more kinds. The above-mentioned light stabilizer is used in a proportion of 0.005 to 5 parts by weight, preferably 0.005 to 2 parts by weight, more preferably 0.01 to 1 part by weight, per 100

parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (A) and the high-pressure-processed low-density polyethylene (B).

[0095]

Specific examples of ultraviolet absorbers include, for example, salicylic acid ultraviolet absorbers, such as phenyl salicylate, p-tert-butylphenyl salicylate and p-octylphenyl salicylate; benzophenone ultraviolet absorbers, such as 2,4-dihydroxybenzophenone, 2-hydroxy-4-methoxybenzophenone, 2-hydroxy-4-octoxybenzophenone, 2-hydroxy-4-dodecyloxybenzophenone, 2,2'-dihydroxy-4-methoxybenzophenone, 2,2'-dihydroxy-4,4'-dimethoxybenzophenone and 2-hydroxy-4-methoxy-5-sulfobenzophenone; benzotriazole ultraviolet absorbers, such as 2-(2'-hydroxy-5'-methylphenyl)benzotriazole, 2-(2'-hydroxy-5'-tert-butylphenyl)benzotriazole, 2-(2'-hydroxy-3',5'-di-tert-butylphenyl)benzotriazole, 2-(2'-hydroxy-3'-tert-butyl-5'-methylphenyl)-5-chlorobenzotriazole, 2-(2'-hydroxy-3',5'-di-tert-butylphenyl)-5-chlorobenzotriazole and 2-(2'-hydroxy-3',5'-di-tert-amylphenyl)benzotriazole; cyanoacrylate ultraviolet absorbers, such as 2-ethylhexyl-2-cyano-3,3'-diphenyl acrylate and ethyl-2-cyano-3,3'-diphenyl acrylate.

[0096]

The above-mentioned ultraviolet absorber is used in a proportion of 0.005 to 5 parts by weight, preferably 0.005 to 2 parts by weight, more preferably 0.01 to 1 part by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (A) and the high-pressure-processed low-density polyethylene (B).

[0097]

The inorganic compound is an inorganic oxide, an inorganic hydroxide, a hydrotalcite or the like, each containing at least one atom selected from Mg, Ca, Al and Si which are effective as a thermo-keeping agent.

[0098]

Specifically, there can be mentioned inorganic oxides, such as  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{MgO}$  and  $\text{CaO}$ ; inorganic hydroxides, such as  $\text{Al}(\text{OH})_3$ ,  $\text{Mg}(\text{OH})_2$  and  $\text{Ca}(\text{OH})_2$ ; inorganic composite compounds represented by the formula:  $\text{M}^{2+}_{1-x}\text{Al}_x(\text{OH})_2(\text{A}^{n-})_{x/n} \cdot m\text{H}_2\text{O}$  (wherein  $\text{M}^{2+}$  represents an ion of a bivalent metal, such as Mg, Ca or Zn;  $\text{A}^{n-}$  represents an anion, such as  $\text{Cl}^-$ ,  $\text{Br}^-$ ,  $\text{I}^-$ ,  $\text{NO}_3^{2-}$ ,  $\text{ClO}_4^-$ ,  $\text{SiO}_4^{2-}$ ,  $\text{CO}_3^{2-}$ ,  $\text{SiO}_3^{2-}$ ,  $\text{HPO}_4^{2-}$ ,  $\text{HBO}_3^{2-}$  and  $\text{PO}_4^{2-}$ ;  $x$  represents a number satisfying the requirement:  $0 < x < 0.5$ ; and  $m$  represents a number satisfying the requirement:  $0 \leq m \leq 2$ ), and hydrotalcite which is a calcination product of the above compounds. Of these, preferred is hydrotalcite, and especially preferred are calcination products of the inorganic composite compounds represented by the above formula.

[0099]

The above-mentioned inorganic compounds can be used independently or in combination. It is desired that the average particle diameter of the inorganic compound is 10  $\mu\text{m}$  or less, preferably 5  $\mu\text{m}$  or less, more preferably 3  $\mu\text{m}$  or less.

[0100]

By using the inorganic compound having an average particle diameter within the above range, a film having an excellent clarity can be obtained. The above-mentioned

inorganic compound is used in a proportion of 1 to 20 parts by weight, preferably 1 to 18 parts by weight, more preferably 2 to 15 part by weight, per 100 parts by weight of the total amount of ethylene- $\alpha$ -olefin copolymer (A) and high-pressure-processed low-density polyethylene (B).

[0101]

When the inorganic compound is used in the above-mentioned proportion, a film having an excellent heat insulating property can be obtained. The agricultural film of the present invention has a layer comprising the above-mentioned ethylene copolymer composition (1).

[0102]

When the agricultural film of the present invention is a single-layer film comprising a layer composed of the above-mentioned ethylene copolymer composition (1), the thickness of this agricultural film is generally 30 to 200  $\mu\text{m}$ , preferably 40 to 180  $\mu\text{m}$ , more preferably 50 to 150  $\mu\text{m}$ .

[0103]

The agricultural film of the present invention which is the above single-layer film has the following physical properties and characteristics.

(i) The Elmendorf tear strength (JIS Z 1702) in the machine direction (MD) (extrusion direction of the film) is 90 kg/cm or more, preferably 100 kg/cm or more, and that in the traverse direction (TD) (width direction of the film) is 90 kg/cm or more, preferably 100 kg/cm or more.

(ii) The dart impact strength (JIS Z 1707) at a thickness of 100  $\mu\text{m}$  is 450 g or more, preferably 500 g or more.

(iii) The tensile strength at break (JIS K 6781) in the MD is 350 kg/cm<sup>2</sup> or more, preferably 400 kg/cm<sup>2</sup> or more, and

that in the TD is 350 kg/cm<sup>2</sup> or more, preferably 400 kg/cm<sup>2</sup> or more.

[0104]

The agricultural film of the present invention having the above-mentioned single-layer structure can be prepared from the above-mentioned ethylene copolymer composition (1) by a film molding, for example, inflation method or T-die method.

[0105]

The film molding by this inflation method is performed by extruding ethylene copolymer composition (1) through a circular die and expanding the extruded composition by a predetermined air flow. It is preferred that the resin temperature during extrusion of ethylene copolymer composition (1) is 190 to 250°C.

[0106]

As mentioned above, the agricultural film of the present invention may have a multi-layer structure having a layer composed of the above-mentioned ethylene copolymer composition (1). As the agricultural film of the present invention having a multi-layer structure, for example, a three-layer laminate film comprising the below-mentioned outer layer, intermediate layer and inner layer is especially preferred.

[0107]

Outer layer

The outer layer constituting the above-mentioned three-layer laminate film is formed of ethylene copolymer composition (2) containing ethylene- $\alpha$ -olefin copolymer (E),

and if desired, high-pressure-processed low-density polyethylene (B), anti-fogging agent (C), and nonionic fluorine-containing surfactant (D).

[0108]

[Ethylene- $\alpha$ -olefin copolymer (E)]

The ethylene- $\alpha$ -olefin copolymer (E) forming the outer layer of the above-mentioned three-layer laminate film is substantially the same as the ethylene- $\alpha$ -olefin copolymer (A) used in the formation of the above-mentioned single-layer agricultural film except that it is a copolymer of ethylene and an  $\alpha$ -olefin having 4 to 12 carbon atoms, and has a density of 0.925 to 0.940 g/cm<sup>3</sup>, preferably 0.925 to 0.935 g/cm<sup>3</sup>.

[0109]

Such ethylene- $\alpha$ -olefin copolymer (E) can be produced by the same method as the production method for the above-mentioned ethylene- $\alpha$ -olefin copolymer (A), namely, by copolymerizing ethylene with an  $\alpha$ -olefin having 4 to 12 carbon atoms in the presence of a single-site catalyst, for example, a metallocene catalyst for olefin polymerization so that the density of the resultant copolymer becomes 0.925 to 0.940 g/cm<sup>3</sup>.

[0110]

In the present invention, the ethylene- $\alpha$ -olefin copolymer (E) is used in a proportion of 60 to 100 parts by weight, preferably 65 to 99 parts by weight, more preferably 70 to 95 parts by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (E) and the high-pressure-processed low-density

polyethylene (B).

[0111]

[High-pressure-processed low-density polyethylene (B)]

The high-pressure-processed low-density polyethylene (B) forming the outer layer of the above-mentioned three-layer laminate film is the same as the high-pressure-processed low-density polyethylene (B) used in the formation of the above-mentioned agricultural film having a single-layer structure.

[0112]

In the present invention, the high-pressure-processed low-density polyethylene (B) is used, if desired, in a proportion of 0 to 40 parts by weight, preferably 1 to 35 parts by weight, more preferably 5 to 30 parts by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (E) and the high-pressure-processed low-density polyethylene (B).

[0113]

[Other components]

In the ethylene copolymer composition (2) forming the above-mentioned outer layer, if desired, the above-mentioned anti-fogging agent (C) and the nonionic fluorine-containing surfactant (D), and an additive, such as the conventionally known weathering agent, antioxidant, inorganic compound, antistatic agent, thermal stabilizer or the like, as long as the desired effects of the present invention are not impaired.

[0114]

The weathering agent used in the formation of the outer layer is the same as the weathering agent

(ultraviolet absorber, light stabilizer) used, if desired, in the formation of the above-mentioned agricultural film having a single-layer structure. The light stabilizer is used in a proportion of 0.005 to 5 parts by weight, preferably 0.005 to 2 parts by weight, more preferably 0.01 to 1 part by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (E) and the high-pressure-processed low-density polyethylene (B). In addition, the ultraviolet absorber is used in a proportion of 0.005 to 5 parts by weight, preferably 0.005 to 2 parts by weight, more preferably 0.01 to 1 part by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (E) and the high-pressure-processed low-density polyethylene (B).

[0115]

The anti-fogging agent (C) used is the same as that used in the formation of the above-mentioned agricultural film having a single-layer structure. The anti-fogging agent (C) is used in a proportion of 0.05 to 5 parts by weight, preferably 0.1 to 4 parts by weight, more preferably 0.5 to 3 parts by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (E) and the high-pressure-processed low-density polyethylene (B).

[0116]

The above-mentioned nonionic fluorine-containing surfactant (D) is the same as that used in the formation of the above-mentioned agricultural film having a single-layer structure. The nonionic fluorine-containing surfactant (D) is used in a proportion of 0.01 to 1 part by weight, preferably 0.05 to 0.3 part by weight, per 100 parts by

weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (E) and the high-pressure-processed low-density polyethylene (B).

[0117]

Further, the inorganic compound used here is the same as that used, if desired, in the formation of the above-mentioned agricultural film having a single-layer structure. The inorganic compound is used in a proportion of 1 to 20 parts by weight, preferably 1 to 18 parts by weight, more preferably 2 to 15 parts by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (E) and the high-pressure-processed low-density polyethylene (B). When the inorganic compound is used in the above-mentioned proportion in the formation of the outer layer, a three-layer laminate film having an excellent heat insulating property can be obtained.

[0118]

Intermediate layer

The intermediate layer constituting the above-mentioned three-layer laminate film is formed of the ethylene copolymer composition (3) containing the ethylene- $\alpha$ -olefin copolymer (F), the anti-fogging agent (C), and, if desired, the high-pressure-processed low-density polyethylene (B) and the nonionic fluorine-containing surfactant (D).

[0119]

[Ethylene- $\alpha$ -olefin copolymer (F)]

The ethylene- $\alpha$ -olefin copolymer (F) forming the intermediate layer of the above-mentioned three-layer laminate film is substantially the same as the above-

mentioned ethylene- $\alpha$ -olefin copolymer (A) except that it is a copolymer of ethylene and an  $\alpha$ -olefin having 4 to 12 carbon atoms, and has a density of 0.880 to 0.920 g/cm<sup>3</sup>, preferably 0.890 to 0.915 g/cm<sup>3</sup>.

[0120]

Such ethylene- $\alpha$ -olefin copolymer (F) can be produced by the same method as the production method for the above-mentioned ethylene- $\alpha$ -olefin copolymer (A), namely, by copolymerizing ethylene with an  $\alpha$ -olefin having 4 to 12 carbon atoms in the presence of a single-site catalyst, for example, a metallocene catalyst for olefin polymerization so that the density of the resultant copolymer becomes 0.880 to 0.920 g/cm<sup>3</sup>.

[0121]

In the present invention, the ethylene- $\alpha$ -olefin copolymer (F) is used in a proportion of 60 to 100 parts by weight, preferably 65 to 99 parts by weight, more preferably 70 to 95 parts by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (F) and the high-pressure-processed low-density polyethylene (B).

[0122]

[High-pressure-processed low-density polyethylene (B)]

High-pressure-processed low-density polyethylene (B) forming the intermediate layer of the above-mentioned three-layer laminate film is the same as the high-pressure-processed low-density polyethylene (B) used in the formation of the above-mentioned agricultural film having a single-layer structure.

[0123]

In the present invention, the high-pressure-processed low-density polyethylene (B) is used in a proportion of 0 to 40 parts by weight, preferably 1 to 35 parts by weight, more preferably 5 to 30 parts by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (F) and the high-pressure-processed low-density polyethylene (B).

) [0124]

[Anti-fogging agent (C)]

The anti-fogging agent (C) added to the ethylene- $\alpha$ -olefin copolymer (F) and the high-pressure-processed low-density polyethylene (B) forming the above-mentioned intermediate layer is the same as the anti-fogging agent (C) used in the formation of the above-mentioned agricultural film having a single-layer structure.

[0125]

The anti-fogging agent (C) is used in a proportion of 0.05 to 5 parts by weight, preferably 0.1 to 4 parts by weight, more preferably 0.5 to 3 parts by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (F) and the high-pressure-processed low-density polyethylene (B).

[0126]

[Other components]

In the ethylene copolymer composition (3) for forming the above-mentioned intermediate layer, if desired, the above-mentioned nonionic fluorine-containing surfactant (D), and an additive, such as the conventionally known weathering agent, antioxidant, inorganic compound,

antistatic agent, thermal stabilizer or the like, as long as the desired effects of the present invention are not impaired.

[0127]

The above-mentioned nonionic fluorine-containing surfactant (D) is the same as the nonionic fluorine-containing surfactant (D) used in the formation of the above-mentioned agricultural film having a single-layer structure. The nonionic fluorine-containing surfactant (D) is used in a proportion of 0.01 to 1 part by weight, preferably 0.05 to 0.3 part by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (F) and the high-pressure-processed low-density polyethylene (B).

[0128]

The weathering agent used in the formation of the intermediate layer is the same as the weathering agent (ultraviolet absorber, light stabilizer) used if desired in the formation of the above-mentioned agricultural film having a single-layer structure. The light stabilizer is used in a proportion of 0.005 to 5 parts by weight, preferably 0.005 to 2 parts by weight, more preferably 0.01 to 1 part by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (F) and the high-pressure-processed low-density polyethylene (B). In addition, the ultraviolet absorber is used in a proportion of 0.005 to 5 parts by weight, preferably 0.005 to 2 parts by weight, more preferably 0.01 to 1 part by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (F) and the high-pressure-processed low-density polyethylene (B).

[0129]

Further, the inorganic compound used in the formation of the intermediate layer is the same as that used if desired in the formation of the above-mentioned agricultural film having a single-layer structure. The inorganic compound is used in a proportion of 1 to 20 parts by weight, preferably 1 to 18 parts by weight, more preferably 2 to 15 parts by weight, per 100 parts by weight of the total amount of the ethylene- $\alpha$ -olefin copolymer (F) and the high-pressure-processed low-density polyethylene (B). When the inorganic compound is used in the above-mentioned proportion in the formation of the intermediate layer, a three-layer laminate film having an excellent heat insulating property can be obtained.

[0130]

Inner layer

The inner layer constituting the above-mentioned three-layer laminate film is formed of the ethylene copolymer composition (1) containing the above-mentioned ethylene- $\alpha$ -olefin copolymer (A), the anti-fogging agent (C), the nonionic fluorine-containing surfactant (D), and if desired, the high-pressure-processed low-density polyethylene (B). Description of this composition (1) was already given.

[0131]

In the present invention, the density ( $d_E$  or  $d_{EB}$ ) of the resin forming the above-mentioned outer layer and the density ( $d_A$  or  $d_{AB}$ ) of the resin forming the inner layer have the relationship satisfying  $d_E$  or  $d_{EB} \geq d_A$  or  $d_{AB}$ . The resin herein means an ethylene- $\alpha$ -olefin copolymer or a

copolymer of an ethylene- $\alpha$ -olefin copolymer and a high-pressure-processed low-density polyethylene.

[0132]

Therefore, the anti-fogging agent (C) contained in the above-mentioned intermediate layer migrates always towards the inner layer rather than towards the outer layer, so that the anti-fogging properties of the inner layer can be maintained.

[0133]

When the density of the ethylene- $\alpha$ -olefin copolymer (A) forming the inner layer is much smaller than 0.905 g/cm<sup>3</sup>, the tackiness of the internal surface of the film is large and the sliding to the pipe house is poor, leading to poor workability of the spreading film.

[0134]

Further, it is desired to use the above-mentioned maleic wax in combination with the anti-fogging agent (C) from the viewpoint of improving the anti-fogging properties maintaining ability. In the agricultural film of the present invention which is the above-mentioned three-layer laminate film, the thickness of the outer layer is generally in the range of from 3 to 100  $\mu$ m, preferably from 10 to 80  $\mu$ m, more preferably from 20 to 70  $\mu$ m, the thickness of the intermediate layer is in the range of from 10 to 150  $\mu$ m, preferably from 20 to 120  $\mu$ m, more preferably from 30 to 100  $\mu$ m, the thickness of the inner layer is in the range of from 3 to 100  $\mu$ m, preferably from 10 to 80  $\mu$ m, more preferably from 20 to 70  $\mu$ m, and the total thickness of all of the above layers is in the range of from 30 to

200  $\mu\text{m}$ , preferably from 50 to 180  $\mu\text{m}$ , more preferably from 70 to 150  $\mu\text{m}$ .

[0135]

In such a three-layer laminate film, it is desired that the thickness ratio between the outer layer [I], the intermediate layer [II] and the inner layer [III] (outer layer [I]/intermediate layer [II]/inner layer [III]) is 0.2 to 4/1 to 10/1, preferably 0.5 to 2/2 to 6/1.

[0136]

The agricultural film of the present invention which is the above-mentioned three-layer laminate film has the following physical properties and characteristics.

- (i) The Elmendorf tear strength (JIS Z 1702) in the MD is 90 kg/cm or more, preferably 100 kg/cm or more, and that in the TD is 90 kg/cm or more, preferably 100 kg/cm or more.
- (ii) The dart impact strength (JIS Z 1707) at a thickness of 100  $\mu\text{m}$  is 900 g or more, preferably 1000 g or more.
- (iii) The tensile strength at break (JIS K 6781) in the MD is 350 kg/cm<sup>2</sup> or more, preferably 370 kg/cm<sup>2</sup> or more, and that in the TD is 350 kg/cm<sup>2</sup> or more, preferably 370 kg/cm<sup>2</sup> or more.

[0137]

The agricultural film of the present invention having the above-mentioned three-layer structure can be prepared by mixing together components including the polyethylene resins used for the respective layers and the above-mentioned additives and the like, and melt-mixing the resultant mixture by a Banbury mixer, a roll mill, an extruder or the like, and then, laminating an outer layer, an intermediate layer and an inner layer by a co-extrusion

inflation method or a co-extrusion T-die method.

[0138]

The agricultural film of the present invention comprising two layers or four or more layers can be prepared in accordance with these preparation methods.

[0139]

[Effect of the Invention]

) The present invention can provide an agricultural film having excellent anti-fogging properties maintaining ability, excellent dust-proof property and excellent toughness, as compared with the above-mentioned conventional agricultural films (single-layer and multi-layer films). Further, the agricultural film of the present invention is lightweight, and has a small content of the low molecular weight component in the polymer used. Therefore, the film has advantages not only in that the tackiness thereof is small, and thus, the workability of spreading films is excellent, but also in that adherence of the films to one another at a high temperature rarely occurs. Particularly, in the agricultural film comprising the above-mentioned three-layer laminate film, the resin forming the outer layer has a density higher than that of the resin forming the inner layer, resulting in very excellent anti-fogging properties maintaining ability (long-term anti-fogging properties).

[0140]

) The agricultural film according to the present invention has the above effects, and therefore, can be used for the plantation of useful plants over a long term by spreading on farm equipment, such as a house, a tunnel and the like.

[0141]

The present invention will be described below with reference to the following Examples, which should not be construed as limiting the scope of the present invention. In the Examples and Comparative Examples, various evaluations for the agricultural films were conducted by the following methods.

(1) Anti-fogging properties

On February 5, 1997, three-layer films having a thickness of 100  $\mu\text{m}$  (outer layer/intermediate layer/inner layer = 20  $\mu\text{m}$ /60  $\mu\text{m}$ /20  $\mu\text{m}$ ) were spread on the pipe house (frontage: 3.6 m; depth: 20 m) provided in the site of Mitsui Chemicals Co., Ltd., Sodegaura-shi, Chiba-ken.

[0142]

Thereafter, the state of drops of water adhering to the internal surface of the film was observed with time every 7 a.m. to 8 a.m.

<5-Rank evaluation>

5: Water is in an anti-stick state, and drops of water are not recognized.

4: Water is in an anti-stick state, but flow lines remain.

3: Both of flow lines and drops of water exist.

2: Both of flow lines and drops of water exist, and more drops of water exist.

1: Fine drops of water adhere to almost entire surface of the film.

A film evaluated as rank 3 or higher passes the anti-fogging properties test, and a film evaluated as rank 1 or 2 does not pass.

[0143]

(2) Elmendorf tear strength

Strength in the vertical direction and the traverse direction was measured by the method in accordance with JIS Z-1702.

(3) Dart impact strength

Dart impact strength was determined by conducting an impact test in accordance with JIS Z 1707 (dart tip diameter: 38 mm).

[0144]

(4) Tensile strength at break

Tensile strength at break is a value determined by conducting a tensile test in accordance with JIS K 6781 under the conditions below with respect to the multi-layer film in the machine direction (MD) and the traverse direction (TD) using a constant crosshead travel rate-type tensile tester (manufactured and sold by Instron Corporation).

[0145]

[Test conditions]

Sample: JIS K 6781

Atmosphere temperature: 23°C

Tensile speed: 500 mm/min

[0146]

[Reference Example 1]

Preparation of ethylene-1-hexene copolymer

[Preparation of olefin polymerization catalyst]

5.0 kg of silica dried at 250°C for 10 hours was suspended in 80 liters of toluene, followed by cooling to 0°C. Then, 28.7 liters of a toluene solution of methylaluminoxane (Al: 1.33 mol/liter) was added thereto

dropwise over 1 hour. In this instance, the temperature of the system was maintained at 0°C. Subsequently, a reaction is conducted at 0°C for 60 minutes, and then, the temperature was elevated to 95°C over 1.5 hours, followed by a reaction at that temperature for 20 hours. Then, the temperature was lowered to 60°C, and the supernatant was removed by decantation.

[0147]

The thus obtained solid component was washed with toluene two times, and then, suspended in 80 liters of toluene again. To the resultant system were added dropwise 7.4 liters of a toluene solution of bis(1,3-n-butylmethylcyclopentadienyl)zirconium dichloride (Zr: 34.0 mmol/liter) and 1.0 liter of a toluene solution of bis(1,3-dimethylcyclopentadienyl)zirconium dichloride (Zr: 28.1 mmol/liter) at 80°C over 30 minutes, and a reaction was conducted at 80°C for another 2 hours. Then, the supernatant was removed and washed with hexane two times, thereby obtaining a solid catalyst containing 3.6 mg of zirconium per 1 g.

[Preparation of prepolymerization catalyst]

To 85 liters of hexane containing 1.7 mol of triisobutylaluminum were added 0.85 kg of the above-obtained solid catalyst and 25 g of 1-hexane, and a prepolymerization of ethylene was conducted at 35°C for 12 hours, thereby obtaining a prepolymerization catalyst having 10 g of polyethylene prepolymerized per 1 g of the solid catalyst. The intrinsic viscosity ( $\eta$ ) of this ethylene polymer was 1.74 dl/g.

[Polymerization]

Using two continuous fluidized bed gas phase polymerization apparatuses connected to each other in series, a copolymerization of ethylene with 1-hexene was conducted in the presence of the above-obtained prepolymerization catalyst, to thereby obtain an ethylene-1-hexene copolymer.

[0148]

The thus obtained ethylene-1-hexene copolymer had a 1-hexene content of 7.5 % by weight, a density of 0.928 g/cm<sup>3</sup>, a melt flow rate (MFR; ASTM D 1238-65T, at 190°C and under a load of 2.16 kg) of 1.63 g/10 minutes, and a molecular weight distribution (Mw/Mn) of 3.5 as measured by gel permeation chromatography (GPC).

[0149]

In addition, with respect to this copolymer, the temperature (T<sub>m</sub>) at the largest peak in the endothermic curve as measured by differential scanning calorimeter (DSC) was 120°C, and the weight percentage (W) of the component soluble in n-decane at room temperature was 0.25 % by weight.

[0150]

With respect to this copolymer, the above-mentioned B<sub>1</sub> as measured by GPC-IR analysis was 12.2/1000C (12.2 per 1000 carbon atoms), and B<sub>2</sub> was 9.9/1000C.

[0151]

[Reference Example 2]

Using two continuous fluidized bed gas phase polymerization apparatuses connected to each other in

series, a copolymerization of ethylene with 1-hexene was conducted in the presence of the above prepolymerization catalyst described in Reference Example 1, to thereby obtain an ethylene-1-hexene copolymer.

[0152]

The thus obtained ethylene-1-hexene copolymer had a 1-hexene content of 11 % by weight, a density of 0.915 g/cm<sup>3</sup>, a melt flow rate (MFR; ASTM D 1238-65T, at 190°C and under a load of 2.16 kg) of 2.19 g/10 minutes, and a molecular weight distribution (Mw/Mn) of 3.5 as measured by GPC.

[0153]

In addition, with respect to this copolymer, the temperature (T<sub>m</sub>) at the largest peak in the endothermic curve as measured by DSC was 108°C, and the weight percentage (W) of the component soluble in n-decane at room temperature was 0.42 % by weight.

[0154]

With respect to this copolymer, the above-mentioned B<sub>1</sub> as measured by GPC-IR analysis was 18.6/1000C, and B<sub>2</sub> was 14.7/1000C.

[0155]

[Reference Example 3]

The ethylene-1-hexene copolymers shown in Table 1 were obtained in the same manner as in Reference Example 1.

[0156]

[Table 1]

Table 1

	Reference example	Density (d) [g/cm <sup>3</sup> ]	MFR [g/10min]	Mw/Mn	W [wt%]	B <sub>1</sub> B <sub>2</sub>   [1/1000C]		Comonomer		Tm [°C]	Swelling ratio (%)
								Kind	Content [wt%]		
Ethylene · α - olefin copolymer (I) (II) (III)	1	0.928	1.63	3.5	0.25	12.2	9.9	1-Hexene	7.5	120	-
	2	0.915	2.19	3.5	0.42	18.6	14.7	1-Hexene	11.0	108	-
	3	0.908	1.95	3.0	0.54	16.7	13.3	1-Hexene	13.5	100	-
High-pressure-processed low-density polyethylene (IV)	-	0.923	0.51	4.0	-	-	-	-	-	-	40

[0157]

[Examples 1 to 8 and Comparative Examples 1 to 6]

Using the resins shown in Table 1 and the anti-fogging agents shown in Table 2, three-layer films having a thickness of 100  $\mu\text{m}$  and compositions shown in Table 3 were prepared by inflation molding under the following conditions.

[Inflation molding conditions]

Molding machine: Three-layer machine, manufactured by Alpine Co., Ltd.

Die bore diameter: 400 mm

Molding temperature: 200°C

Flat width: 1500 mm

Thickness: All of layers: 100  $\mu\text{m}$  (outer layer/intermediate layer/inner layer = 20  $\mu\text{m}$ /60  $\mu\text{m}$ /20  $\mu\text{m}$ )

The physical properties of the thus obtained films are shown in Table 4.

[0158]

Table 2

Table 2

[illegible]

Note) Glycerol monostearate: Trade name S-100, Riken vitamin

**Sorbital monostearate:** Trade name S-300, Riken vitamin

monodistearate: Marubishi Yuka

**Sorbitan monodistearate E03: Marubishi Yuka**

Sorbitan monodistearate E05: Marubishi Yuka

Sorbitan monodistearate E08: Marubishi Yuka

**E020: Trade name T-60, Matsumoto oil & fat pharmaceutical**

monodistearate E03: Marubishi Yuka

monodistearate E05: Marubishi Yuka

E03, E05, E08 and E020 each represent the case where n in the formula  $(-\text{CH}_2\text{CH}_2\text{O}-)_n\text{H}$  is 3, 5, 8 and 20, respectively.

[0159]

Table 3

Outer layer		(I) / (IV)
Composition for resin		= 85 / 15
Density of resin mixture	[g/cm <sup>3</sup> ]	0. 927
Anti-fogging agent	[wt%]	1. 0
Weathering agent	[wt%]	0. 2
Hydrotalcite	[wt%]	3. 0
Thickness of film	[μm]	2 0
Intermediate layer		(II) / (IV)
Composition for resin		= 85 / 15
Density of resin mixture	[g/cm <sup>3</sup> ]	0. 916
Anti-fogging agent	[wt%]	2. 0
Weathering agent	[wt%]	0. 2
Hydrotalcite	[wt%]	7. 0
Fluorine-containing surfactant	[wt%]	0. 1
Thickness of film	[μm]	6 0
Inner layer		(III) / (IV)
Composition for resin		= 85 / 15
Density of resin mixture	[g/cm <sup>3</sup> ]	0. 910
Anti-fogging agent	[wt%]	2. 0
Maleic acid-modified polyethylene		
Weathering agent	[wt%]	0. 1
Hydrotalcite	[wt%]	0
Fluorine-containing surfactant	[wt%]	0. 1
Thickness of film	[μm]	2 0

[0160]

(Note 1) Weathering agent: N,N'-bis(3-aminopropyl)ethylenediamine-2-4-bis[N-butyl-N-(1,2,2,6,6-pentamethyl-4-piperidyl)amino]-6-chloro-1,3,5-triazine condensation product

(Note 2) Hydrotalcite: DHT-4A (trade name; manufactured by Kyowa Chemical Industry Co., Ltd.)

(Note 3) Fluorine-containing surfactant: DS-403 (trade name; manufactured DAIKIN INDUSTRIES, Ltd.)

[0161]

[Table 4]

[0162]  
[Table 5]

Table 4

	Comparative Example 1	Comparative Example 2	Example 1	Example 2	Example 3	Comparative Example 3
<Anti-fogging properties>						
Initial value	3	4	5	5	5	5
After 1 month	3	4	5	5	5	5
After 2 month	3	4	5	5	5	4
After 8 month	3	3	5	5	4	4
After 9 month	3	3	5	5	4	4
After 10 month	3	3	5	4	4	3
After 11 month	3	2	4	4	4	3
After 12 month	2	2	4	4	3	2
After 13 month	2	2	4	4	3	2
Elmendorf tear strength						
MD/TD [kgf/cm]	140/230	150/230	140/230	150/230	150/240	120/210
Dart impact strength [g]	>1700	>1700	>1700	>1700	>1700	>1700
Tensile strength at break						
MD/TD [kg/cm <sup>2</sup> ]	480/490	480/490	490/500	500/510	500/510	440/450

(Note) In Comparative Example 3, during inflation molding, the amount of the resin extruded was unstable. Further, the obtained film was discolored brown.

Table 4 (Continued)

	Example 4	Example 5	Comparative Example 4	Example 6	Example 7	Comparative Example 5	Comparative Example 6	Example 8
<Anti-fogging properties>								
Initial value	5	5	5	5	5	5	4	5
After 1 month	5	5	5	5	5	5	3	5
After 2 month	5	5	5	5	5	4	3	4
After 8 month	5	5	5	4	4	4	3	4
After 9 month	5	4	4	4	4	3	3	4
After 10 month	4	4	3	4	4	3	3	4
After 11 month	4	3	3	4	3	2	3	3
After 12 month	4	3	2	3	3	2	2	3
After 13 month	4	3	2	3	3	2	2	3
Elmendorf tear strength								
MD/TD [kgf/cm]	140/240	140/230	140/230	150/230	140/220	140/210	150/240	140/230
Dart impact strength [g]	>1700	>1700	>1700	>1700	>1700	>1700	>1700	>1700
Tensile strength at break								
MD/TD [kg/cm <sup>2</sup> ]	490/490	480/480	480/490	480/480	460/480	440/460	510/520	490/490

(Note) In Comparative Example 5, during inflation molding, the amount of the resin extruded was unstable.

[0163]

As described above, by adding a nonionic fluorine-containing surfactant having a fluorine content in a specific range and a surface tension as the aqueous solution thereof in a specific range, the anti-fogging properties level and the anti-fogging properties maintaining ability are improved.